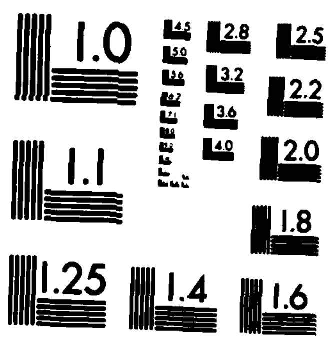
AD-A146 636 THEORY OF HARMONIC GENERATION OF FINITE AMPLITUDE ULTRASONIC WAVES IN SOL..(U) TENNESSEE UNIV KNOXVILLE DEPT OF PHYSICS AND ASTRONOMY J PHILIP ET AL. OCT 84 1/1

UNCLASSIFIED TR-22 N00014-81-K-0229 F/G 12/1 NL



'OPY RESOLUTION TEST CHART

12

DEPARTMENT OF PHYSICS AND ASTRONOMY

OFFICE OF NAVAL RESEARCH
CONTRACT NO. N00014-81-K-0229
PROJECT NO. 384-306
TECHNICAL REPORT NO. 22

THEORY OF HARMONIC GENERATION OF FINITE AMPLITUDE ULTRASONIC WAVES IN SOLIDS OF HEXAGONAL SYMMETRY

by

Jacob Philip and M. A. Breazeale

UNIVERSITY OF TENNESSEE

KNOXVILLE
TN 37996-1200

OFFICE OF NAVAL RESEARCH
CONTRACT NO. N00014-81-K-0229
PROJECT NO. 384-306
TECHNICAL REPORT NO. 22

THEORY OF HARMONIC GENERATION OF FINITE AMPLITUDE ULTRASONIC WAVES IN SOLIDS OF HEXAGONAL SYMMETRY

by

Jacob Philip and M. A. Breazeale

M. A. Breazeale, Principal Investigator
Ultrasonics Laboratory
Department of Physics
The University of Tennessee
Knoxville, Tennessee 37996-1200

October 1984

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER Technical Report No. 22	2. GOVT ACCESSION NO. AD-A146636	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) Theory of Harmonic Generation of Finite Amplitude Ultrasonic Waves in Solids of Hexagonal Symmetry		5. TYPE OF REPORT & PERIOD COVERED Interim
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Jacob Philip and M. A. Breazeale		8. CONTRACT OR GRANT NUMBER(s) N00014-81-K-0229
9. PERFORMING ORGANIZATION NAME AND ADDRESS Department of Physics University of Tennessee Knoxville, TN 37996-1200		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61153N; RR011-08-01; NR 384-306
11. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research, Code 412 Department of the Navy Arlington, VA 22217		12. REPORT DATE October 1984
		13. NUMBER OF PAGES 61
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

nonlinearity parameter
ultrasonic harmonic generation
nonlinear hexagonal crystal
third-order elastic constants

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The theory of the propagation of a finite amplitude ultrasonic wave along pure mode directions in a nonlinear crystal of hexagonal symmetry is the subject of the technical report. Beginning with the Brugger thermodynamic definition of the second-order elastic (SOE) and the third-order elastic (TOE) constants, one can derive the strain energy for a generalized Lagrangian strain. By introducing the appropriate rotation matrices and the stress tensor one can write the equations of motion in the Lagrangian coordinate

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE
S/N 0102-LF-014-6601

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

Block 20 continued:

frame. Nonlinear differential wave equations are derived for waves propagating along the symmetry axis and along any direction in the basal plane. Solutions are given for describing the growth of the second harmonic of an initially sinusoidal ultrasonic wave in the directions considered, so that one can determine TOE constants from harmonic generation experiments. A unique representation is given of the variation of the nonlinearity parameter with angle for all directions in the basal plane, and it is concluded that from harmonic generation experiments one should be able to measure three combinations of the ten TOE constants: $(2C_{222} - C_{112})$, $\frac{1}{2}(3C_{112} - 4C_{111})$, and C_{333}. Possible other combinations are to be presented in a future technical report.

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

PREFACE

The theory of the propagation of a finite amplitude wave in isotropic solids and crystals of cubic symmetry was begun a number of years ago [M. A. Breazeale and Joseph Ford, J. Appl. Phys. <u>36</u>, 3486-3490 (1965)], and subsequently was developed in terms of the elegant Lagrangian formalism [Albert C. Holt and Joseph Ford, J. Appl. Phys. <u>40</u>, 142-148 (1969)]. This theory has served as the basis of measurement of the third-order elastic (TOE) constants of many isotropic solids (e.g., John H. Cantrell, Jr., Technical Report No. 14) and crystals of cubic symmetry (e.g., Jacob Philip. Technical Report No. 19). For isotropic solids three TOE constants are necessary, and for cubic crystals one finds six. However, many materials of technological importance have lower symmetry than cubic. Therefore, it now is desirable to determine which TOE constants enter into harmonic generation in waves of finite amplitude propagating in crystals of lower symmetry. This Technical Report is the first effort in this direction.

The Technical Report gives a complete analysis of the theory of propagation of a finite amplitude ultrasonic wave in a or near crystal of hexagonal symmetry along the symmetry axis and along any direction in the basal plane. From the theory we conclude that at least four of the ten ~~TOE~~ third order elastic constants can be evaluated from harmonic generation measurements in the appropriate directions in hexagonal crystals.

Completion of the Technical Report was made possible by the efforts of Gonghuan Du whose meticulous mathematical efforts are gratefully acknowledged.

CHAPTER I

INTRODUCTION

Although the nonlinear acoustical properties of fluids has been studied for more than a century, a systematic study of the nonlinear acoustical properties of solids actually began about three decades ago. Work in this area gained momentum rapidly because a large number of the basic properties of solids can be explained only on the basis of a nonlinear theory. Among these properties are thermal expansion, the interaction of lattice waves, inequality of adiabatic and isothermal elastic constants, and the dependence of the elastic constants on pressure and temperature.

As is well known, the description of the nonlinear properties of solids is made in terms of the higher-order elastic constants. These constants, coefficients of the higher-order terms in the relation between internal energy and lattice strains on a solid, are some of the most important parameters in the description of the nonlinear properties of solids. Their measurement, therefore, has attracted considerable attention. The ordinary elastic constants, of course, are determined directly from the sound velocity. They are the second-order elastic constants. The third-order elastic constants are measured by techniques outlined in a previous Technical Report.[1] As was pointed out in that Report, most of the measurements reported are on solids of cubic symmetry with six third-order elastic (TOE) constants. As the symmetry is lowered, the number of elastic constants increases.[2] For example,

crystals with hexagonal symmetry have 10 TOE constants and those with trigonal symmetry have 14 (or 20) TOE constants.

Only very few measurements have been reported on the TOE constants of hexagonal and trigonal crystals, in part because of a lack of a general theory. The linear theory of elastic wave propagation along different directions in these types of media has been given by Musgrave,[3] Farnell,[4] and others. A complete description of the linear theory has been given by Musgrave.[5] The purpose of the present study is to expand on the linear theory to include nonlinear terms and to formulate the theory in such a way that the nonlinear distortion of finite amplitude ultrasonic waves in hexagonal crystals can be described.

In an anisotropic medium there are only certain directions along which elastic waves can propagate as the pure longitudinal modes which are of primary interest in the application of the harmonic generation technique. Associated with any propagation direction there are three independent waves, the displacements of which form a mutually orthogonal set. In general, none of the three displacement vectors coincides with the vector which is normal to the wavefront; i.e., in general, the waves are neither pure longitudinal nor pure transverse. The specific directions in which pure mode longitudinal waves propagate have been determined by Borgnis[6] for crystals with trigonal, hexagonal, tetragonal and cubic symmetry and general conditions have been established under which pure longitudinal waves exist. In order to determine the third-

order elastic constants from wave speed measurements in stressed crystals, it is necessary to know these pure mode directions.

The theory of harmonic generation of longitudinal waves, which is discussed in detail in later chapters, also depends upon availability of pure longitudinal mode directions. Using the method due to Borgnis,[6] Brugger[7] has determined the pure mode directions for all crystal point groups belonging to orthorhombic, tetragonal, cubic, trigonal (rhombohedral) and hexagonal systems. Also Brugger[7] has solved the eigenvalue problem for each of these directions and polarization vectors and the wave speeds have been tabulated. A similar description for transverse waves has been given by Chang.[8]

The theory of finite deformations and its application to crystalline solids including nonlinear terms has been given by Murnaghan.[9] The theory has been applied to cubic crystals subjected to finite hydrostatic compression by Birch[10] and later by Seeger and Buck.[11] Einspruch and Manning[12] extended the theory to hexagonal, tetragonal and orthorhombic symmetry. Thurston and Brugger[13,14] solved the equations of small amplitude waves in a homogeneously deformed crystal and expressed their results in terms of the "natural velocity" and its stress derivative. They defined the "natural velocity" as the length of the specimen in the unstressed state divided by the wave travel time in the stressed state. They presented results for isotropic and cubic solids. The necessary relations between third-order elastic coefficients and stress derivatives of sound speeds along different directions in crystals of orthorhombic, tetragonal, cubic, trigonal and

hexagonal classes have been derived by Brugger[15] using his thermodynamic definition of third-order elastic constants.[16]

The TOE constants of a large number of crystals belonging to cubic symmetry have been measured. Most of these measurements involve measurement of the stress derivatives of ultrasonic wave velocities in solids. The first measurement of this kind was by Lazarus[17] on KCl, NaCl, CuZn, Cu and Al. Later Hughes and Kelly[18] made measurements on polystyrene, iron and pyrex glass and reported the three Murnaghan TOE constants of these materials. The first measurement of the complete set of six TOE constants of a cubic solid was by Bateman et al.[19] on germanium. Later, measurements have been reported on many cubic crystals by several workers. The values reported up to 1979 have been tabulated by Hearmon.[2]

The second most widely used technique to measure the TOE constants of solids is the ultrasonic harmonic generation technique developed by Breazeale and Ford.[20] Measurements have been reported on a number of solids.[21] The theory of second harmonic generation in cubic crystals and the experimental technique has been described in detail in a previous report.[1] Also for a review of the work done on the TOE constants of cubic crystals, refer to Hearmon[2] and Green.[22]

Even though the TOE constants of a number of isotropic and cubic crystals have been determined, not many measurements have been reported on crystals of lower symmetry. In all the work reported on such crystals so far, the hydrostatic and uniaxial stress derivatives of ultrasonic wave velocities have been measured and the TOE constants have been determined from those data. Since the number of TOE constants

for crystals of lower symmetry is higher, a correspondingly higher number of careful measurements are to be made to isolate all the TOE constants. In the following paragraphs we outline the measurements to be made on hexagonal crystals with 10 TOE constants. The values measured until 1979 on these solids also have been tabulated by Hearmon.[2]

The complete set of ten TOE constants of the hexagonal crystal zinc has been determined from measurements of the hydrostatic and uniaxial stress dependence of ultrasonic wave velocity by Swartz and Elbaum.[23] They used data from twelve measurements for various propagation, polarization and stress application directions to isolate all the 10 TOE constants. An interference technique has been used by them to measure the wave velocity difference in stressed and unstressed samples. They have reported the temperature and pressure derivatives of the SOE constants in their work. The Grüneisen parameter γ of zinc has been evaluated by the authors and the volume thermal expansion coefficient calculated from that is in agreement with experimental value of the volume thermal expansion coefficient.

The complete set of the ten TOE constants of the hexagonal metal magnesium has been determined by Naimon,[24] who measured the hydrostatic pressure and uniaxial compression derivatives of natural sound velocities using ultrasonic pulse-superposition technique. With the sample oriented with faces perpendicular to the a, b, c axes, four hydrostatic and ten uniaxial experiments for different propagation and polarization directions were used to determine the ten TOE constants. Even though the hydrostatic pressure derivatives of the SOE constants of beryllium,[25]

zirconium,[26] europium,[27] rhenium,[28] magnesium,[29] cadmium,[30], gadolenium,[31] dysprosium,[31] and titanium[31] have been reported, that information is not sufficient to determine the complete set of TOE constants. The TOE constants of a number of hexagonal solids have been determined theoretically by a number of workers using the method of homogeneous deformations with different model potentials. For a review of the theoretical work, refer to Ramji Rao and Ramanand.[32]

In the following report we develop the nonlinear theory required to consistently describe the propagation of an initially sinusoidal ultrasonic wave along pure mode directions in a solid of hexagonal symmetry. As pointed out by Brugger,[7] pure mode directions in a hexagonal crystal are (1) the symmetry axis, (2) all directions in the basal plane, and (3) directions along a cone centered on the symmetry axis, the apex angle of the cone depending on the SOE constants of the hexagonal crystal under consideration. In the following report we specialize the equations to the pure mode directions (1) and (2) and show which TOE constants can be measured by propagating finite amplitude waves along these directions. We end the report with some comments about the way in which the theory can be handled for directions (3) along a cone centered on the symmetry axis.

CHAPTER II

GENERAL THEORY OF NONLINEAR WAVE PROPAGATION IN SOLIDS

Consider a point P in the medium with coordinates a_i (a,b,c) in the unstrained state. Let the point P move to P' with coordinates x_i (x,y,z) in the deformed state. The components of the displacement can then be written as

$$
\begin{aligned}
U &= x - a \\
V &= y - b \qquad (1) \\
W &= z - c
\end{aligned}
$$

In the theory of finite deformations of an elastic solid due to the large deformations involved, the initial coordinates of a particle in the undeformed state are not interchangeable with the final coordinates in the deformed state. The expression for the strain energy in terms of the strains change correspondingly. In the Lagrangian formulation, the strain is described in the initial or undeformed state and the initial coordinates of the material particle a_i are taken as independent variables. The Lagrangian formulation is used in the theory described in this technical report.

The Lagrangian strain parameters which are components of the finite strain tensor are given by[9]

$$\eta = \frac{1}{2} (J^*J - \delta) \qquad (2)$$

where J is the Jacobian matrix given by

$$J = \begin{pmatrix} \frac{\partial x}{\partial a} & \frac{\partial x}{\partial b} & \frac{\partial x}{\partial c} \\ \frac{\partial y}{\partial a} & \frac{\partial y}{\partial b} & \frac{\partial y}{\partial c} \\ \frac{\partial z}{\partial a} & \frac{\partial z}{\partial b} & \frac{\partial z}{\partial c} \end{pmatrix} = \begin{pmatrix} 1 + \frac{\partial U}{\partial a} & \frac{\partial U}{\partial b} & \frac{\partial U}{\partial c} \\ \frac{\partial V}{\partial a} & 1 + \frac{\partial V}{\partial b} & \frac{\partial V}{\partial c} \\ \frac{\partial W}{\partial a} & \frac{\partial W}{\partial b} & 1 + \frac{\partial W}{\partial c} \end{pmatrix} \tag{3}$$

If we write $\frac{\partial U}{\partial a} = U_a$, $\frac{\partial V}{\partial b} = V_b$, etc.

$$J = \begin{pmatrix} 1 + U_a & U_b & U_c \\ V_a & 1 + V_b & V_c \\ W_a & W_b & 1 + W_c \end{pmatrix} \tag{4}$$

J^* is the transpose of J given by

$$J^* = \begin{pmatrix} 1 + U_a & V_a & W_a \\ U_b & 1 + V_b & W_b \\ U_c & V_c & 1 + W_c \end{pmatrix} \tag{5}$$

and

$$\delta = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{6}$$

is the unit matrix. The stress-strain relation is given by[9]

$$\sigma = (\rho/\rho_0)\ J\ (\frac{\partial \phi}{\partial \eta}) J^* \tag{7}$$

where σ is the Cauchy stress tensor and ρ_0 and ρ are the densities in the undeformed and deformed state, respectively. ϕ is the strain energy per unit undeformed volume and

$$\frac{\partial\phi}{\partial\eta} = \begin{bmatrix} \frac{\partial\phi}{\partial\eta_{11}} & \frac{\partial\phi}{\partial\eta_{12}} & \frac{\partial\phi}{\partial\eta_{13}} \\ \frac{\partial\phi}{\partial\eta_{21}} & \frac{\partial\phi}{\partial\eta_{22}} & \frac{\partial\phi}{\partial\eta_{23}} \\ \frac{\partial\phi}{\partial\eta_{31}} & \frac{\partial\phi}{\partial\eta_{32}} & \frac{\partial\phi}{\partial\eta_{33}} \end{bmatrix} \tag{8}$$

The properties of the crystalline medium enter into the theory through the strain energy term ϕ. Many deformable media with crystalline structure are elastically insensitive to certain prerotations depending upon the symmetry of the medium. For those rotations to which the medium is insensitive we have the relation

$$\phi(R^*\eta R) = \phi(\eta) \tag{9}$$

where R is the rotation matrix and R* is the transpose of R. ϕ can be expanded and written as a sum of terms of different degrees in the elements of η as follows:

$$\phi = \phi_0 + \phi_1 + \phi_2 + \phi_3 + \ldots \tag{10}$$

In that case the series relation

$$\phi_j(R^*\eta R) = \phi_j(\eta) \tag{11}$$

holds good, ϕ being homogeneous functions of degree j in the elements of η, j = 0,1,2,3 ... In terms of the elastic moduli, Eq. (10) can be written as:

$$\phi = \phi_0 + k_1 C_{ij} \eta_{ij} + k_2 C_{ijk\ell} \eta_{ij} \eta_{k\ell}$$

$$+ k_3 C_{ijk\ell mn} \eta_{ij} \eta_{k\ell} \eta_{mn} + \cdots \tag{12}$$

where the C's are the elastic constants and k_n is a constant factor depending on the definition of elastic constants. If the initial energy and deformation of the body are neglected, the first two terms in (12) are zero, and

$$\phi = k_2 C_{ijk\ell} \eta_{ij} \eta_{k\ell} + k_3 C_{ijk\ell mn} \eta_{ij} \eta_{k\ell} \eta_{mn} + \cdots \tag{13}$$

As is well known, $C_{ijk\ell}$ are the second-order and $C_{ijk\ell mn}$ are the third-order elastic constants. In accordance with Brugger's thermodynamic definition of elastic constants,[16] the factor k_n is $\frac{1}{n!}$. So, based on that definition we have $k_2 = \frac{1}{2}$ and $k_3 = \frac{1}{6}$. Using single subscript notation instead of double subscript notation for elastic constants and using Brugger's definition of elastic constants, the general expression for ϕ can be written as

$$\phi = \frac{1}{2} C_{\lambda\mu} \eta_{ij} \eta_{k\ell} + \frac{1}{6} C_{\lambda\mu\nu} \eta_{ij} \eta_{k\ell} \eta_{mn} + \text{higher order terms} \tag{14}$$

where $\lambda \rightarrow ij$, $\mu \rightarrow k\ell$ and $\nu \rightarrow mn$. The second-order stiffness $C_{\lambda\mu}$ form a fourth rank tensor containing 81 components, of which 21 are independent for the most unsymmetric triclinic and the third-order stiffness form a sixth rank tensor with 729 components of which 56 are independent for a triclinic crystal. The number of elastic constants decreases considerably for crystals of higher symmetry.

Equation (14) for the strain energy takes appropriate forms for crystals of different symmetry. The number of elastic constants in different crystal classes have been worked out by various authors and have been presented in a tabular form by Hearmon[2] which in conjunction with Eq. (14) conveniently can be used to obtain an expression for the strain energy of any class of crystal.

The principle of conservation of mass along with the definition of J leads to the relation

$$|J| = \rho_0/\rho \simeq (1 + U_a + V_b + W_c) . \qquad (15)$$

The equations of motion for an elastic medium are restatements of Newton's second law. For convenience one can introduce the stress tensor T, which is not symmetric, as

$$T = J(\frac{\partial \phi}{\partial \eta}) \qquad (16)$$

with nine components. This expression allows one to write the equations of motion as

$$\frac{\partial T_{ij}}{\partial a_j} = \rho_0 \ddot{U}_i \qquad (17)$$

in the Lagrangian coordinate frame.* These equations of motion take

*In the Eulerian coordinate system the corresponding equations of motion are $\frac{\partial \sigma_{ij}}{\partial x_j} = \rho \ddot{U}_i$

where σ_{ij} is the Cauchy stress tensor which is symmetric and x_j are the coordinates of the particle λ in the deformed state.

particular forms along the axes. Along the a, b and c axes they are

$$
\begin{aligned}
\rho_0 \ddot{U} &= \partial T_{11}/\partial a + \partial T_{12}/\partial b + \partial T_{13}/\partial c \\
\rho_0 \ddot{V} &= \partial T_{21}/\partial a + \partial T_{22}/\partial b + \partial T_{23}/\partial c \\
\rho_0 \ddot{W} &= \partial T_{31}/\partial a + \partial T_{32}/\partial b + \partial T_{33}/\partial c
\end{aligned}
\tag{18}
$$

From Eqs. (4), (8) and (16), the stress matrix T can be written in component form as

$$
\begin{pmatrix} T_{11} & T_{12} & T_{13} \\ T_{21} & T_{22} & T_{23} \\ T_{31} & T_{32} & T_{33} \end{pmatrix} = \begin{pmatrix} J_{11} & J_{12} & J_{13} \\ J_{21} & J_{22} & J_{23} \\ J_{31} & J_{32} & J_{33} \end{pmatrix} \begin{pmatrix} \frac{\partial\phi}{\partial\eta_{11}} & \frac{\partial\phi}{\partial\eta_{12}} & \frac{\partial\phi}{\partial\eta_{13}} \\ \frac{\partial\phi}{\partial\eta_{21}} & \frac{\partial\phi}{\partial\eta_{22}} & \frac{\partial\phi}{\partial\eta_{23}} \\ \frac{\partial\phi}{\partial\eta_{31}} & \frac{\partial\phi}{\partial\eta_{32}} & \frac{\partial\phi}{\partial\eta_{33}} \end{pmatrix}
\tag{19}
$$

From this we can write down the components as follows:

$$
\left.
\begin{aligned}
T_{11} &= J_{11} \frac{\partial\phi}{\partial\eta_{11}} + J_{12} \frac{\partial\phi}{\partial\eta_{21}} + J_{13} \frac{\partial\phi}{\partial\eta_{31}} \\
T_{12} &= J_{11} \frac{\partial\phi}{\partial\eta_{12}} + J_{12} \frac{\partial\phi}{\partial\eta_{22}} + J_{13} \frac{\partial\phi}{\partial\eta_{32}} \\
T_{13} &= J_{11} \frac{\partial\phi}{\partial\eta_{13}} + J_{12} \frac{\partial\phi}{\partial\eta_{23}} + J_{13} \frac{\partial\phi}{\partial\eta_{33}} \\
T_{21} &= J_{21} \frac{\partial\phi}{\partial\eta_{11}} + J_{22} \frac{\partial\phi}{\partial\eta_{21}} + J_{23} \frac{\partial\phi}{\partial\eta_{31}} \\
T_{22} &= J_{21} \frac{\partial\phi}{\partial\eta_{12}} + J_{22} \frac{\partial\phi}{\partial\eta_{22}} + J_{23} \frac{\partial\phi}{\partial\eta_{32}} \\
T_{23} &= J_{21} \frac{\partial\phi}{\partial\eta_{13}} + J_{22} \frac{\partial\phi}{\partial\eta_{23}} + J_{23} \frac{\partial\phi}{\partial\eta_{33}} \\
T_{31} &= J_{31} \frac{\partial\phi}{\partial\eta_{11}} + J_{32} \frac{\partial\phi}{\partial\eta_{21}} + J_{33} \frac{\partial\phi}{\partial\eta_{31}} \\
T_{32} &= J_{31} \frac{\partial\phi}{\partial\eta_{12}} + J_{32} \frac{\partial\phi}{\partial\eta_{22}} + J_{33} \frac{\partial\phi}{\partial\eta_{32}} \\
T_{33} &= J_{31} \frac{\partial\phi}{\partial\eta_{13}} + J_{32} \frac{\partial\phi}{\partial\eta_{23}} + J_{33} \frac{\partial\phi}{\partial\eta_{33}}
\end{aligned}
\right\}
\tag{20}
$$

Note that the T_{ij} tensor defined here is not symmetric.

We will consider the case of plane finite amplitude waves propagating along the axes of the medium under consideration. For plane waves propagating along the a-axis the displacement components become

$$\begin{aligned} U &= U(a,t) \\ V &= V(a,t) \\ W &= W(a,t) \end{aligned} \tag{21}$$

For this special case, the equations of motion given by Eq. (18) become

$$\begin{aligned} \rho_0 \ddot{U} &= \partial T_{11}/\partial a \\ \rho_0 \ddot{V} &= \partial T_{21}/\partial a \\ \rho_0 \ddot{W} &= \partial T_{31}/\partial a \end{aligned} \tag{22}$$

Similarly for plane waves propagating along the b-axis, we have

$$\begin{aligned} \rho_0 \ddot{U} &= \partial T_{12}/\partial b \\ \rho_0 \ddot{V} &= \partial T_{22}/\partial b \\ \rho_0 \ddot{W} &= \partial T_{32}/\partial b \end{aligned} \tag{23}$$

and for plane waves propagating along the c-axis, we have

$$\begin{aligned} \rho_0 \ddot{U} &= \partial T_{13}/\partial c \\ \rho_0 \ddot{V} &= \partial T_{23}/\partial c \\ \rho_0 \ddot{W} &= \partial T_{33}/\partial c \end{aligned} \tag{24}$$

These equations are written out in more explicit terms and solved for crystals belonging to hexagonal symmetry in the following chapters. The elastic strain energy appropriate to the crystal symmetry is obtained. Only terms up to third order are retained in the strain energy expression as we are interested only in the first order of nonlinearity which is enough to account for most of the nonlinear properties. Moreover, inclusion of fourth- and higher-order terms make the algebra unwieldy.

CHAPTER III

SECOND HARMONIC GENERATION OF ULTRASOUND IN CRYSTALS OF HEXAGONAL SYMMETRY

There are two classes of crystals with hexagonal symmetry, one with the Hermann-Mauguin symbol 6, $\bar{6}$, 6/m with 5 second-order and 12 third-order elastic constants and the other with the symbol 622, 6mm, $\bar{6}$m2, 6/mmm with 5 second-order and 10 third-order elastic constants. Since most of the crystals with hexagonal symmetry belong to the second class, we will confine our attention to it. The figure of the hexagonal close packed structure is shown in Fig. 1. A drawing of the coordinate frame and the basal plane are also given in the figure. The five SOE constants are C_{11}, C_{12}, C_{13}, C_{33} and C_{44} and the ten TOE constants are C_{111}, C_{112}, C_{113}, C_{123}, C_{133}, C_{144}, C_{155}, C_{222}, C_{333} and C_{344}.[2]

As has been shown by Borgnis[6] and by Brugger,[7] any axis in the xy plane and the z-axis are directions in which pure mode longitudinal wave propagation is possible (in the linear approximation). To be consistent with the general theory in Chapter II, the x-direction is represented as a-axis and the 60° rotated direction from the x-direction is designated as a'-axis. The z-direction and c-axis coincide.

Using Brugger's definition of third order elastic constants, expression for the strain energy density for hexagonal crystals is given by:

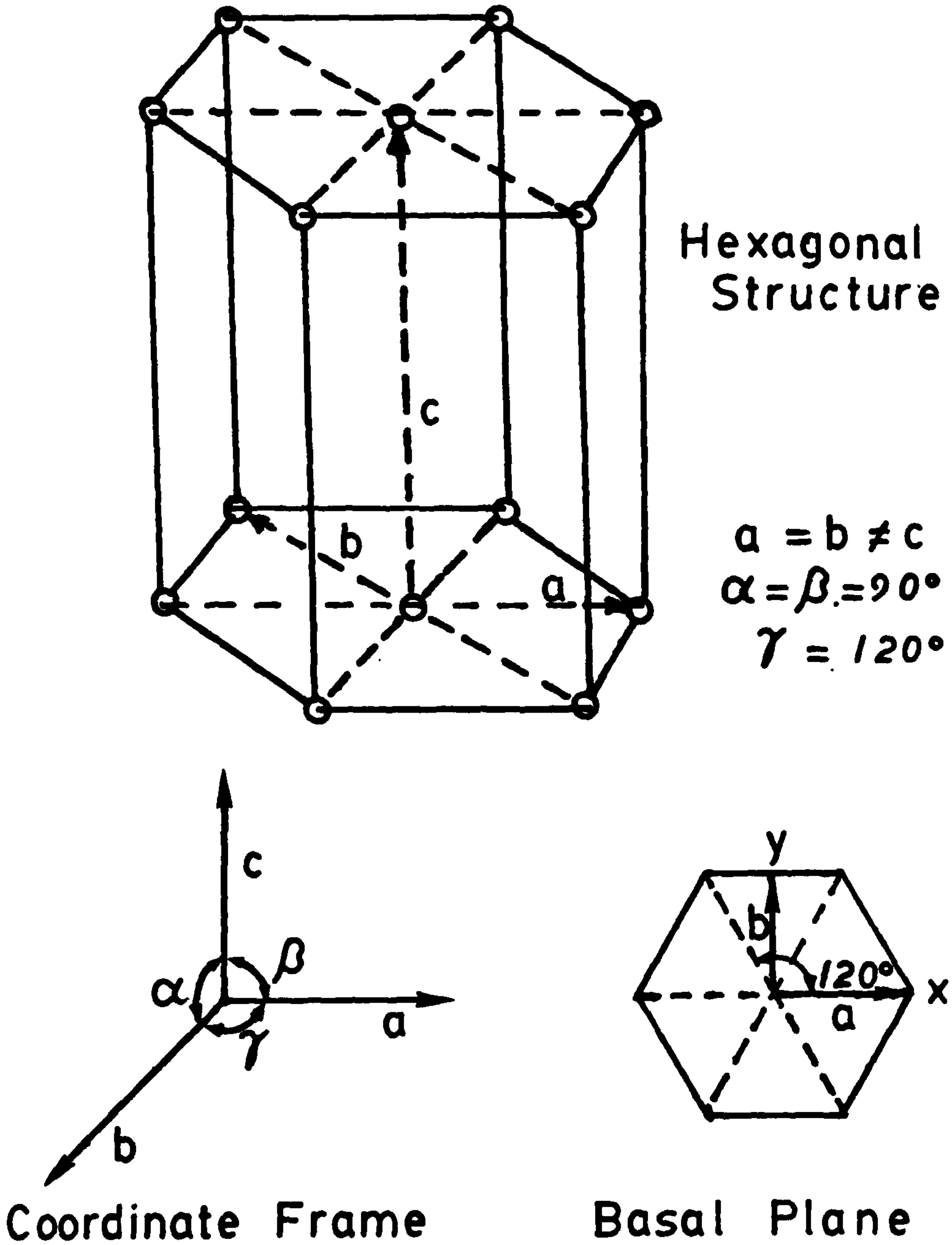


Fig. 1. Hexagonal close packed structure.

$$\begin{aligned}
\phi = {} & \frac{1}{2} C_{11}(\eta_{11}{}^2 + \eta_{22}{}^2 + \eta_{12}{}^2 + \eta_{21}{}^2) \\
& + C_{12}(\eta_{11}\eta_{22} - \frac{1}{2}\eta_{12}{}^2 - \frac{1}{2}\eta_{21}{}^2) \\
& + C_{13}(\eta_{11}\eta_{33} + \eta_{22}\eta_{33}) + \frac{1}{2} C_{33}\eta_{33}{}^2 \\
& + C_{44}(\eta_{13}{}^2 + \eta_{31}{}^2 + \eta_{23}{}^2 + \eta_{32}{}^2) \\
& + \frac{1}{6} C_{111}(\eta_{11} + \eta_{22})^3 \\
& + \frac{1}{2} C_{113}\eta_{33}(\eta_{11} + \eta_{22})^2 + \frac{1}{2} C_{133}\eta_{33}{}^2(\eta_{11} + \eta_{22}) \\
& + C_{144}(\eta_{11} + \eta_{22})(\eta_{23}{}^2 + \eta_{32}{}^2 + \eta_{13}{}^2 + \eta_{31}{}^2) \\
& + \frac{1}{2} C_{166}(\eta_{11} + \eta_{22})[(\eta_{11} - \eta_{22})^2 + 2(\eta_{12}{}^2 + \eta_{21}{}^2)] \\
& + \frac{1}{6} C_{266}[(\eta_{11} - \eta_{22})^3 - 6(\eta_{12}{}^2 + \eta_{21}{}^2)(\eta_{11} - \eta_{22})] \\
& + \frac{1}{6} C_{333}\eta_{33}{}^3 + C_{344}\eta_{33}(\eta_{23}{}^2 + \eta_{32}{}^2 + \eta_{13}{}^2 + \eta_{31}{}^2) \\
& + \frac{1}{2} C_{366}\eta_{33}[(\eta_{11} - \eta_{22})^2 + 2(\eta_{12}{}^2 + \eta_{21}{}^2)] \\
& + 2C_{456}[(\eta_{22} - \eta_{11})(\eta_{23}{}^2 + \eta_{32}{}^2 - \eta_{13}{}^2 - \eta_{31}{}^2) + 4(\eta_{12}\eta_{23}\eta_{31} \\
& \quad + \eta_{21}\eta_{32}\eta_{13})] \, . \qquad (25)
\end{aligned}$$

In this expression C_{166}, C_{266}, C_{366} and C_{456} are combinations of TOE constants given by

$$\begin{aligned}
C_{166} &= \frac{1}{4}(-2C_{111} - C_{112} + 3C_{222}) \\
C_{266} &= \frac{1}{4}(2C_{111} - C_{112} - C_{222}) \\
C_{366} &= \frac{1}{2}(C_{113} - C_{123}) \\
C_{456} &= \frac{1}{2}(-C_{144} + C_{155})
\end{aligned} \qquad (26)$$

Einspruch and Manning[12] derived the above expression using Birch's definition of TOE constants. That expression can easily be converted into Eq. (25) which was Brugger's definition. The relation between Brich and Brugger TOE constants can be obtained from the relation

$$C_{\lambda\mu\nu}^{(\mathrm{Birch})} = M\, C_{\lambda\mu\nu}^{(\mathrm{Brugger})}/6 \tag{27}$$

where M is the possible number of ways in which $C_{\lambda\mu\nu}$ can be expressed in tensor notation.

The strain derivatives of the elastic energy can be obtained by differentiating Eq. (25) with respect to the nine components of the strain parameter. They are given by

$$\begin{aligned}\frac{\partial\phi}{\partial\eta_{11}} = {} & C_{11}\eta_{11} + C_{12}\eta_{22} + C_{13}\eta_{33} + \frac{1}{2}C_{111}\eta_{11}^{2} + C_{111}\eta_{11}\eta_{22} + \frac{1}{2}C_{111}\eta_{22}^{2} \\ & + \frac{1}{2}C_{133}\eta_{33}^{2} + C_{113}\eta_{11}\eta_{33} + C_{113}\eta_{22}\eta_{33} \\ & + C_{144}\eta_{23}^{2} + C_{144}\eta_{32}^{2} + C_{144}\eta_{13}^{2} + C_{144}\eta_{31}^{2} + \frac{3}{2}C_{166}\eta_{11}^{2} \\ & + \frac{1}{2}C_{166}\eta_{22}^{2} - C_{166}\eta_{11}\eta_{22} + C_{166}\eta_{12}^{2} + C_{166}\eta_{21}^{2} \\ & + \frac{1}{2}C_{266}\eta_{11}^{2} - C_{266}\eta_{11}\eta_{22} + \frac{1}{2}C_{266}\eta_{22}^{2} - C_{266}\eta_{12}^{12} \\ & - C_{266}\eta_{21}^{2} + C_{366}\eta_{11}\eta_{33} - C_{366}\eta_{22}\eta_{33} - 2C_{456}\eta_{23}^{2} - 2C_{456}\eta_{32}^{2} \\ & + 2C_{456}\eta_{13}^{2} + 2C_{456}\eta_{31}^{2}\ .\end{aligned} \tag{28a}$$

$$\begin{aligned}\frac{\partial\phi}{\partial\eta_{21}} = {} & C_{11}\eta_{21} - C_{12}\eta_{21} + 2C_{166}\eta_{11}\eta_{21} + 2C_{166}\eta_{22}\eta_{21} - 2C_{266}\eta_{11}\eta_{21} \\ & + 2C_{266}\eta_{22}\eta_{21} + 2C_{366}\eta_{33}\eta_{21} + 8C_{456}\eta_{32}\eta_{13}\ .\end{aligned} \tag{28b}$$

$$\frac{\partial\phi}{\partial\eta_{31}} = 2C_{44}\eta_{31} + 2C_{144}\eta_{11}\eta_{31} + 2C_{144}\eta_{22}\eta_{31} + 2C_{344}\eta_{33}\eta_{31} - 4C_{456}\eta_{22}\eta_{31}$$

$$+ 4C_{456}\eta_{11}\eta_{31} + 8C_{456}\eta_{12}\eta_{23} \,. \tag{28c}$$

$$\frac{\partial\phi}{\partial\eta_{12}} = C_{11}\eta_{12} - C_{12}\eta_{12} + 2C_{166}\eta_{11}\eta_{12} + 2C_{166}\eta_{22}\eta_{12} - 2C_{266}\eta_{11}\eta_{12}$$

$$+ 2C_{266}\eta_{22}\eta_{12} + 2C_{366}\eta_{33}\eta_{12} + 8C_{456}\eta_{23}\eta_{31} \,. \tag{28d}$$

$$\frac{\partial\phi}{\partial\eta_{22}} = C_{11}\eta_{22} + C_{12}\eta_{11} + C_{13}\eta_{33} + \frac{1}{2}C_{111}\eta_{11}{}^2 + C_{111}\eta_{11}\eta_{22}$$

$$+ \frac{1}{2}C_{111}\eta_{22}{}^2 + \frac{1}{2}C_{133}\eta_{33}{}^2 + C_{113}\eta_{11}\eta_{33} + C_{113}\eta_{22}\eta_{33}$$

$$+ C_{144}\eta_{23}{}^2 + C_{144}\eta_{32}{}^2 + C_{144}\eta_{13}{}^2 + C_{144}\eta_{31}{}^2$$

$$- \frac{1}{2}C_{166}\eta_{11}{}^2 + \frac{3}{2}C_{166}\eta_{22}{}^2 - C_{166}\eta_{11}\eta_{22} + C_{166}\eta_{12}{}^2$$

$$+ C_{166}\eta_{21}{}^2 - \frac{1}{2}C_{266}\eta_{11}{}^2 + C_{266}\eta_{11}\eta_{22} - \frac{1}{2}C_{266}\eta_{22}{}^2$$

$$+ C_{266}\eta_{12}{}^2 + C_{266}\eta_{21}{}^2 + C_{366}\eta_{22}\eta_{33} - C_{366}\eta_{11}\eta_{33} + 2C_{456}\eta_{23}{}^2$$

$$+ 2C_{456}\eta_{32}{}^2 - 2C_{456}\eta_{13}{}^2 - 2C_{456}\eta_{31}{}^2 \,. \tag{28e}$$

$$\frac{\partial\phi}{\partial\eta_{32}} = 2C_{44}\eta_{32} + 2C_{144}\eta_{11}\eta_{32} + 2C_{144}\eta_{22}\eta_{32} + 2C_{344}\eta_{33}\eta_{32} + 4C_{456}\eta_{22}\eta_{32}$$

$$- 4C_{456}\eta_{11}\eta_{32} + 8C_{456}\eta_{21}\eta_{13} \,. \tag{28f}$$

$$\frac{\partial\phi}{\partial\eta_{13}} = 2C_{44}\eta_{13} + 2C_{144}\eta_{11}\eta_{13} + 2C_{144}\eta_{22}\eta_{13} + 2C_{344}\eta_{33}\eta_{13} - 4C_{456}\eta_{22}\eta_{13}$$

$$+ 4C_{456}\eta_{11}\eta_{13} + 8C_{456}\eta_{21}\eta_{32} \,. \tag{28g}$$

$$\frac{\partial\phi}{\partial\eta_{23}} = 2C_{44}\eta_{23} + 2C_{144}\eta_{11}\eta_{23} + 2C_{144}\eta_{22}\eta_{23} + 2C_{344}\eta_{33}\eta_{23} + 4C_{456}\eta_{22}\eta_{23}$$
$$- 4C_{456}\eta_{11}\eta_{23} + 8C_{456}\eta_{12}\eta_{31} \tag{28h}$$

$$\frac{\partial\phi}{\partial\eta_{33}} = C_{13}\eta_{11} + C_{13}\eta_{22} + C_{33}\eta_{33} + \frac{1}{2}C_{333}\eta_{33}^2 + C_{133}\eta_{11}\eta_{33} + C_{133}\eta_{22}\eta_{33}$$
$$+ \frac{1}{2}C_{113}\eta_{11}^2 + C_{113}\eta_{11}\eta_{22} + \frac{1}{2}C_{113}\eta_{22}^2 + C_{344}\eta_{23}^2 + C_{344}\eta_{32}^2$$
$$+ C_{344}\eta_{13}^2 + C_{344}\eta_{31}^2 + \frac{1}{2}C_{366}\eta_{11}^2 + \frac{1}{2}C_{366}\eta_{22}^2 - C_{366}\eta_{11}\eta_{22}$$
$$+ C_{366}\eta_{12}^2 + C_{366}\eta_{21}^2 . \tag{28i}$$

To derive expressions for T_{ij}, the $\frac{\partial\phi}{\partial\eta_{ij}}$ given by Eqs. (28) and the components of the Jacobian matrix are required. The components of the Jacobian matrix in terms of the displacement gradients are (from Eq. (4)):

$$\begin{aligned}
J_{11} &= 1 + U_a = 1 + \partial U/\partial a \\
J_{12} &= U_b = \partial U/\partial b \\
J_{13} &= U_c = \partial U/\partial c \\
J_{21} &= V_a = \partial V/\partial a \\
J_{22} &= 1 + V_b = 1 + \partial V/\partial b \\
J_{23} &= V_c = \partial V/\partial c \\
J_{31} &= W_a = \partial W/\partial a \\
J_{32} &= W_b = \partial W/\partial b \\
J_{33} &= 1 + W_c = 1 + \partial W/\partial c
\end{aligned} \tag{29}$$

The strain parameters can be expressed in terms of displacement gradients if we substitute (4), (5) and (6) into (2). Substitution and simplification give the following expressions for η_{ij}

$$
\begin{aligned}
\eta_{11} &= U_a + \tfrac{1}{2}(U_a^2 + V_a^2 + W_a^2) \\
\eta_{12} = \eta_{21} &= \tfrac{1}{2}(U_b + V_a + U_aU_b + V_aV_b + W_aW_b) \\
\eta_{13} = \eta_{31} &= \tfrac{1}{2}(U_c + W_a + U_aU_c + V_aV_c + W_aW_c) \\
\eta_{22} &= V_b + \tfrac{1}{2}(U_b^2 + V_b^2 + W_b^2) \\
\eta_{23} = \eta_{32} &= \tfrac{1}{2}(V_c + W_b + U_bU_c + V_bV_c + W_bW_c) \\
\eta_{33} &= W_c + \tfrac{1}{2}(U_c^2 + V_c^2 + W_c^2)
\end{aligned}
\tag{30}
$$

To evaluate all the T_{ij}'s, we need the quadratic terms also in η_{ij}; *i.e., we have to evaluate* η_{11}^2, $\eta_{11}\eta_{12}$, *etc. Retaining terms* only up to the quadratic, we get the following expressions for these terms.

$$\eta_{11}^2 = U_a^2 \tag{31a}$$

$$\eta_{22}^2 = V_b^2 \tag{31b}$$

$$\eta_{33}^2 = W_c^2 \tag{31c}$$

$$\eta_{11}\eta_{22} = U_aV_b \tag{31d}$$

$$\eta_{11}\eta_{33} = U_aW_c \tag{31e}$$

$$\eta_{22}\eta_{33} = V_bW_c \tag{31f}$$

$$\eta_{12}^2 = \eta_{21}^2 = \tfrac{1}{4}(U_b^2 + V_a^2 + 2U_bV_a) \tag{31g}$$

$$\eta_{13}{}^2 = \eta_{31}{}^2 = \frac{1}{4}(U_c{}^2 + W_a{}^2 + 2U_cW_a) \tag{31h}$$

$$\eta_{23}{}^2 = \eta_{32}{}^2 = \frac{1}{4}(V_c{}^2 + W_b{}^2 + 2V_cW_b) \tag{31i}$$

$$\eta_{11}\eta_{21} = \eta_{11}\eta_{12} = \frac{1}{2}(U_aU_b + U_aV_a) \tag{31j}$$

$$\eta_{11}\eta_{13} = \eta_{11}\eta_{31} = \frac{1}{2}(U_aU_c + U_aW_a) \tag{31k}$$

$$\eta_{11}\eta_{23} = \eta_{11}\eta_{32} = \frac{1}{2}(U_aV_c + U_aW_b) \tag{31ℓ}$$

$$\eta_{22}\eta_{12} = \eta_{22}\eta_{21} = \frac{1}{2}(V_bU_b + V_aV_b) \tag{31m}$$

$$\eta_{22}\eta_{13} = \eta_{22}\eta_{31} = \frac{1}{2}(V_bU_c + V_bW_a) \tag{31n}$$

$$\eta_{22}\eta_{32} = \eta_{22}\eta_{23} = \frac{1}{2}(V_bV_c + V_bW_b) \tag{31o}$$

$$\eta_{33}\eta_{12} = \eta_{33}\eta_{21} = \frac{1}{2}(W_cU_b + W_cV_a) \tag{31p}$$

$$\eta_{33}\eta_{13} = \eta_{33}\eta_{31} = \frac{1}{2}(W_cU_c + W_aW_c) \tag{31q}$$

$$\eta_{33}\eta_{23} = \eta_{33}\eta_{32} = \frac{1}{2}(W_cV_c + W_cW_b) \tag{31r}$$

$$\begin{aligned}\eta_{12}\eta_{13} &= \eta_{21}\eta_{13} = \eta_{21}\eta_{31} = \eta_{12}\eta_{31} \\ &= \frac{1}{4}(U_bU_c + U_bW_a + V_aV_c + V_aW_a)\end{aligned} \tag{31s}$$

$$\begin{aligned}\eta_{12}\eta_{23} &= \eta_{12}\eta_{32} = \eta_{21}\eta_{23} = \eta_{21}\eta_{32} \\ &= \frac{1}{4}(U_bV_c + U_bW_b + V_aV_c + V_aW_b)\end{aligned} \tag{31t}$$

$$\begin{aligned}\eta_{13}\eta_{23} &= \eta_{13}\eta_{32} = \eta_{31}\eta_{23} = \eta_{31}\eta_{32} \\ &= \frac{1}{4}(U_cV_c + U_cW_b + W_aV_c + W_aW_b)\end{aligned} \tag{31u}$$

The strain parameters are expressed in terms of the displacement gradients by the two sets of Eqs. (30) and Eqs. (31). Now, these expressions are to be substituted into the components of the strain derivatives of the elastic energy given by the set of Eqs. (28). The resulting expressions and the components of the Jacobian matrix given by Eqs. (29) are substituted into the set of Eqs. (20) to obtain the expressions for the components of the stress tensor. After simplification, the components of the stress tensor are:

$$
\begin{aligned}
T_{11} = {} & U_a \cdot C_{11} + V_b \cdot C_{12} + W_c \cdot C_{13} \\
& + U_a{}^2(\tfrac{3}{2} C_{11} + \tfrac{1}{2} C_{111} + \tfrac{3}{2} C_{166} + \tfrac{1}{2} C_{266}) \\
& + V_a{}^2(\tfrac{1}{2} C_{11} + \tfrac{1}{2} C_{166} - \tfrac{1}{2} C_{266}) \\
& + W_a{}^2(\tfrac{1}{2} C_{11} + \tfrac{1}{2} C_{144} + C_{456}) \\
& + U_b{}^2(\tfrac{1}{2} C_{11} + \tfrac{1}{2} C_{166} - \tfrac{1}{2} C_{266}) \\
& + V_b{}^2(\tfrac{1}{2} C_{12} + \tfrac{1}{2} C_{111} - \tfrac{1}{2} C_{166} + \tfrac{1}{2} C_{266}) \\
& + W_b{}^2(\tfrac{1}{2} C_{12} + \tfrac{1}{2} C_{144} - C_{456}) \\
& + U_c{}^2(\tfrac{1}{2} C_{13} + C_{44} + \tfrac{1}{2} C_{144} + C_{456}) \\
& + V_c{}^2(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{144} - C_{456}) \\
& + W_c{}^2(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{133}) \\
& + U_a V_b(C_{12} + C_{111} - C_{166} - C_{266}) \\
& + U_a W_c(C_{13} - C_{113} + C_{366})
\end{aligned}
$$

$$+ V_b W_c (C_{113} - C_{366}) + V_c W_b (C_{144} - 2C_{456})$$

$$+ U_c W_a (C_{44} + C_{144} + 2C_{456}) + U_b V_a (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + C_{166} - C_{266}) \tag{32a}$$

$$T_{12} = U_b \cdot (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12}) + V_a \cdot (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12})$$

$$+ U_a U_b (C_{11} + C_{166} - C_{266})$$

$$+ V_a V_b (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + C_{166} + C_{266})$$

$$+ W_a W_b (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + 2C_{456})$$

$$+ U_a V_a (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + C_{166} - C_{266})$$

$$+ U_b V_b (C_{11} + C_{166} + C_{266})$$

$$+ U_c V_c (C_{44} + 2C_{456})$$

$$+ U_b W_c (C_{13} + C_{366})$$

$$+ V_a W_c \cdot C_{366} + W_a V_c \cdot 2C_{456}$$

$$+ U_c W_b (C_{44} + 2C_{456}) \tag{32b}$$

$$T_{13} = U_c \cdot C_{44} + W_a \cdot C_{44}$$

$$+ U_a U_c (2C_{44} + C_{13} + C_{144} + 2C_{456})$$

$$+ V_a V_c (C_{44} + 2C_{456}) + W_a W_c (C_{44} + C_{344})$$

$$+ U_a W_a (C_{44} + C_{144} + 2C_{456})$$

$$+ V_b U_c (C_{13} + C_{144} -. 2C_{456})$$

$$+ V_bW_a(C_{144} - 2C_{456}) + U_cW_c(C_{33} + C_{344})$$

$$+ U_bV_c(C_{44} + 2C_{456})$$

$$+ U_bW_b(C_{44} + 2C_{456}) + V_aW_b \cdot 2C_{456} \tag{32c}$$

$$T_{21} = U_b(\frac{1}{2}C_{11} - \frac{1}{2}C_{12}) + V_a \cdot (\frac{1}{2}C_{11} - \frac{1}{2}C_{12})$$

$$+ U_aV_a(C_{11} + C_{166} - C_{266})$$

$$+ U_aU_b(\frac{1}{2}C_{11} - \frac{1}{2}C_{12} + C_{166} - C_{266})$$

$$+ V_aV_b(C_{11} + C_{166} + C_{266})$$

$$+ V_aW_c(C_{13} + C_{366})$$

$$+ W_aW_b(\frac{1}{2}C_{11} - \frac{1}{2}C_{12} + 2C_{456})$$

$$+ U_bV_b(\frac{1}{2}C_{11} - \frac{1}{2}C_{12} + C_{166} + C_{266})$$

$$+ U_bW_c \cdot C_{366} + U_cW_b \cdot 2C_{456}$$

$$+ U_cV_c(C_{44} + 2C_{456})$$

$$+ V_cW_a(C_{44} + 2C_{456}) \tag{32d}$$

$$T_{22} = U_a \cdot C_{12} + V_b \cdot C_{11} + W_c \cdot C_{13}$$

$$+ U_a^2(\frac{1}{2}C_{12} + \frac{1}{2}C_{111} - \frac{1}{2}C_{166} - C_{266})$$

$$+ U_b^2(\frac{1}{2}C_{11} + \frac{1}{2}C_{166} + \frac{1}{2}C_{266})$$

$$+ U_c^2(\frac{1}{2}C_{13} + \frac{1}{2}C_{144} - C_{456})$$

$$+ V_a^{\,2}(\tfrac{1}{2} C_{11} + \tfrac{1}{2} C_{166} + \tfrac{1}{2} C_{266})$$

$$+ V_b^{\,2}(\tfrac{3}{2} C_{11} + \tfrac{1}{2} C_{111} + \tfrac{3}{2} C_{166} - \tfrac{1}{2} C_{266})$$

$$+ V_c^{\,2}(\tfrac{1}{2} C_{13} + C_{44} + \tfrac{1}{2} C_{144} + C_{456})$$

$$+ W_a^{\,2}(\tfrac{1}{2} C_{12} + \tfrac{1}{2} C_{144} - C_{456})$$

$$+ W_b^{\,2}(\tfrac{1}{2} C_{11} + \tfrac{1}{2} C_{144} + C_{456})$$

$$+ W_c^{\,2}(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{133}) + U_b V_a(\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + C_{166} + C_{266})$$

$$+ U_a W_c(C_{113} - C_{366}) \quad V_b W_c(C_{13} + C_{113} + C_{366})$$

$$+ U_a V_b(C_{12} + C_{111} - C_{166} + C_{266})$$

$$+ V_c W_b(C_{44} + C_{144} + 2C_{456})$$

$$+ U_c W_a(C_{144} - 2C_{456}) \tag{32e}$$

$$T_{23} = V_c \cdot C_{44} + W_b \cdot C_{44}$$

$$+ U_c V_a(C_{44} + 2C_{456})$$

$$+ V_a W_a(C_{44} + 2C_{456})$$

$$+ U_b U_c(C_{44} + 2C_{456})$$

$$+ V_b V_c(2C_{44} + C_{13} + C_{144} + 2C_{456})$$

$$+ W_b W_c(C_{44} + C_{344})$$

$$+ U_a V_c(C_{13} + C_{144} - 2C_{456})$$

$$+ U_a W_b(C_{144} - 2C_{456})$$

$$+ V_b W_b (C_{44} + C_{144} + 2C_{456})$$

$$+ V_c W_c (C_{33} + C_{344})$$

$$+ U_b W_a \cdot 2C_{456} \tag{32f}$$

$$T_{31} = U_c \cdot C_{44} + W_a \cdot C_{44}$$

$$+ U_a W_a (C_{11} + C_{144} + 2C_{456})$$

$$+ V_b W_a (C_{12} + C_{144} - 2C_{456})$$

$$+ W_a W_c (C_{13} + 2C_{44} + C_{344})$$

$$+ U_b W_b (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + 2C_{456})$$

$$+ V_a W_b (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + 2C_{456})$$

$$+ U_a U_c (C_{44} + C_{144} + 2C_{456})$$

$$+ V_a V_c (C_{44} + 2C_{456})$$

$$+ V_b U_c (C_{144} - 2C_{456})$$

$$+ U_c W_c (C_{44} + C_{344})$$

$$+ U_b V_c \cdot 2C_{456} \tag{32g}$$

$$T_{32} = V_c \cdot C_{44} + W_b \cdot C_{44}$$

$$+ U_b W_a (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + 2C_{456})$$

$$+ V_a W_a (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + 2C_{456})$$

$$+ V_b W_b (C_{11} + C_{144} + 2C_{456})$$

$$+ U_a W_b (C_{12} + C_{144} - 2C_{456})$$

$$+ W_b W_c (C_{13} + 2C_{44} + C_{344})$$

$$+ U_b U_c (C_{44} + 2C_{456})$$

$$+ V_b V_c (C_{44} + C_{144} + 2C_{456})$$

$$+ U_a V_c (C_{144} - 2C_{456})$$

$$+ V_c W_c (C_{44} + C_{344})$$

$$+ V_a U_c \cdot 2C_{456} \qquad (32h)$$

$$T_{33} = U_a \cdot C_{13} + V_b \cdot C_{13} + W_c \cdot C_{33}$$

$$+ U_a^{\,2}(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{113} + \tfrac{1}{2} C_{366})$$

$$+ U_b^{\,2}(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{366})$$

$$+ U_c^{\,2}(\tfrac{1}{2} C_{33} + \tfrac{1}{2} C_{344})$$

$$+ V_a^{\,2}(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{366})$$

$$+ V_b^{\,2}(\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{113} + \tfrac{1}{2} C_{366})$$

$$+ V_c^{\,2}(\tfrac{1}{2} C_{33} + \tfrac{1}{2} C_{344})$$

$$+ W_a^{\,2}(C_{44} + \tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{344})$$

$$+ W_b^{\,2}(C_{44} + \tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{344})$$

$$+ W_c^{\,2}(\tfrac{3}{2} C_{33} + \tfrac{1}{2} C_{333}) + U_c W_a(C_{44} + C_{344})$$

$$+ V_c W_b(C_{44} + C_{344}) + U_a W_c(C_{13} + C_{133})$$

$$+ V_b W_c(C_{13} + C_{133}) + U_a V_b(C_{113} - C_{366}) + U_b V_a(C_{366}) \qquad (32i)$$

A. Longitudinal Wave Propagation along the [100] Direction (or a-Axis)

We intend to propagate only longitudinal waves for two compelling reasons. First, the capacitive receiver used in the measurements responds only to longitudinal displacements; second, the transverse modes are always coupled to the longitudinal modes as is evident from the expressions for the stress tensor given by (32). If we calculate the transverse displacement derivative of the T_{ij} given by (32) along any direction, we find that in the nonlinear terms the longitudinal components always contribute to transverse waves; i.e., transverse waves never propagate as pure modes. This is in agreement with the theory of Goldberg[33] on isotropic solids. On the other hand, longitudinal waves propagate as pure modes in certain directions and are coupled only to their second harmonics in these directions.

For plane longitudinal waves propagating in the [100] direction, the displacement components become

$$U = U(a,t), \qquad V = V(a,t), \qquad W = W(a,t)$$

or U_b, U_c, V_b, V_c, W_b and W_c vanish. So for this case the set of equations (32) simplifies as follows.

$$T_{11} = C_{11}U_a + (\frac{3}{2} C_{11} + \frac{1}{2} C_{111} + \frac{3}{2} C_{166} + \frac{1}{2} C_{266})U_a^2$$
$$+ (\frac{1}{2} C_{11} + \frac{1}{2} C_{166} - \frac{1}{2} C_{266})V_a^2$$
$$+ (\frac{1}{2} C_{11} + \frac{1}{2} C_{144} + C_{456})W_a^2 \quad (33a)$$

$$T_{12} = \frac{1}{2}(C_{11} - C_{12})V_a + \frac{1}{2}(C_{11} - C_{12} + 2C_{166} - 2C_{266})U_aV_a \quad (33b)$$

$$T_{13} = C_{44} \cdot W_a + (C_{44} + C_{144} + 2C_{456})U_aW_a \quad (33c)$$

$$T_{21} = \frac{1}{2}(C_{11} - C_{12})V_a + (C_{11} + C_{166} - C_{266})U_aV_a \quad (33d)$$

$$T_{22} = C_{12}U_a + (\frac{1}{2} C_{12} + \frac{1}{2} C_{111} - \frac{1}{2} C_{166} - C_{266})U_a^2$$
$$+ (\frac{1}{2} C_{11} + \frac{1}{2} C_{166} + \frac{1}{2} C_{266})V_a^2$$
$$+ (\frac{1}{2} C_{12} + \frac{1}{2} C_{144} - C_{456})W_a^2 \quad (33e)$$

$$T_{23} = (C_{44} + 2C_{456})V_aW_a \quad (33f)$$

$$T_{31} = C_{44}W_a + (C_{11} + C_{144} + 2C_{456})U_aW_a \quad (33g)$$

$$T_{32} = (\frac{1}{2} C_{11} - \frac{1}{2} C_{12} + 2C_{456})V_aW_a \quad (33h)$$

$$T_{33} = C_{13}U_a + (\frac{1}{2} C_{13} + \frac{1}{2} C_{113} + \frac{1}{2} C_{366})U_a^2$$
$$+ (\frac{1}{2} C_{13} + \frac{1}{2} C_{366})V_a^2$$
$$+ (C_{44} + \frac{1}{2} C_{13} + \frac{1}{2} C_{344})W_a^2 \quad (33i)$$

The equations of motion for plane wave propagation in the [100] direction are

$$\rho_0 \ddot{U} = \partial T_{11}/\partial a$$

$$\rho_0 \ddot{V} = \partial T_{21}/\partial a \qquad (34)$$

$$\rho_0 \ddot{W} = \partial T_{31}/\partial a$$

Performing the differentiation and substituting for $\partial T_{11}/\partial a$, etc., we get

$$\begin{aligned}\rho_0 \ddot{U} = {} & C_{11}U_{aa} \\ & + (3C_{11} + C_{111} + 3C_{166} + C_{266})U_aU_{aa} \\ & + (C_{11} + C_{166} - C_{266})V_aV_{aa} \\ & + (C_{11} + C_{144} + 2C_{456})W_aW_{aa}\end{aligned} \qquad (35a)$$

$$\rho_0 \ddot{V} = \tfrac{1}{2}(C_{11} - C_{12})V_{aa} + (C_{11} + C_{166} - C_{266})(U_aV_{aa} + V_aU_{aa}) \qquad (35b)$$

$$\rho_0 \ddot{W} = C_{44} \cdot W_{aa} + (C_{11} + C_{144} + 2C_{456})(U_aW_{aa} + W_aU_{aa}) \qquad (35c)$$

where

$$\ddot{U} = \frac{\partial^2 u}{\partial t^2}, \text{ etc.}$$

$$U_a = \frac{\partial u}{\partial a}, \text{ etc.}$$

$$U_{aa} = \frac{\partial^2 u}{\partial a^2}, \text{ etc.}$$

For longitudinal waves along the a-axis we have

$V = 0$ and $W = 0$.

So the equation of motion for this case reduces to

$$\rho_0 \frac{\partial^2 u}{\partial t^2} = C_{11} \frac{\partial^2 u}{\partial a^2}$$

$$+ (3C_{11} + C_{111} + 3C_{166} + C_{266}) \frac{\partial u}{\partial a} \cdot \frac{\partial^2 u}{\partial a^2} \qquad (36)$$

or

$$\rho_0 \frac{\partial^2 u}{\partial t^2} - \alpha \frac{\partial^2 u}{\partial a^2} = \delta \frac{\partial u}{\partial a} \frac{\partial^2 u}{\partial a^2} \qquad (37)$$

where

$$\alpha = C_{11}$$

and

$$\delta = 3C_{11} + C_{111} + 3C_{166} + C_{266} \; .$$

B. Longitudinal Wave Propagation along the [010] Direction

For this case the only nonvanishing displacement components are

$$U = U(b,t), \quad V = V(b,t), \quad W = W(b,t) \; .$$

So for this case the T_{ij}'s given by Eqs. (32) simplify to the following.

$$T_{11} = C_{12}V_b + (\tfrac{1}{2} C_{11} + \tfrac{1}{2} C_{166} - \tfrac{1}{2} C_{266})U_b^{\,2}$$

$$+ (\tfrac{1}{2} C_{12} + \tfrac{1}{2} C_{111} - \tfrac{1}{2} C_{166} + \tfrac{1}{2} C_{266})V_b^{\,2}$$

$$+ (\tfrac{1}{2} C_{12} + \tfrac{1}{2} C_{144} - C_{456})W_b^{\,2} \qquad (38a)$$

$$T_{12} = \tfrac{1}{2}(C_{11} - C_{12})U_b + (C_{11} + C_{166} + C_{266})U_bV_b \qquad (38b)$$

$$T_{13} = (C_{44} + 2C_{456})U_bW_b \qquad (38c)$$

$$T_{21} = \tfrac{1}{2}(C_{11} - C_{12})U_b + (\tfrac{1}{2} C_{11} - \tfrac{1}{2} C_{12} + C_{166} + C_{266})U_bV_b \qquad (38d)$$

$$T_{22} = C_{11}V_b + (\frac{1}{2}C_{11} + \frac{1}{2}C_{166} + \frac{1}{2}C_{266})U_b^{\,2}$$

$$+ (\frac{3}{2}C_{11} + \frac{1}{2}C_{111} + \frac{3}{2}C_{166} - \frac{1}{2}C_{266})V_b^{\,2}$$

$$+ (\frac{1}{2}C_{11} + \frac{1}{2}C_{144} + C_{456})W_b^{\,2} \qquad (38e)$$

$$T_{23} = C_{44} \cdot W_b + (C_{44} + C_{144} + 2C_{456})V_bW_b \qquad (38f)$$

$$T_{31} = (\frac{1}{2}C_{11} - \frac{1}{2}C_{12} + 2C_{456})U_bW_b \qquad (38g)$$

$$T_{32} = C_{44}W_b + (C_{11} + C_{144} + 2C_{456})V_bW_b \qquad (38h)$$

$$T_{33} = C_{13}V_b + (\frac{1}{2}C_{13} + \frac{1}{2}C_{366})U_b^{\,2}$$

$$+ (\frac{1}{2}C_{13} + \frac{1}{2}C_{113} + \frac{1}{2}C_{366})V_b^{\,2}$$

$$+ (\frac{1}{2}C_{13} + C_{44} + C_{344})W_b^{\,2} \,. \qquad (38i)$$

The equations of motion for plane wave propagation in the [010] direction are

$$\rho\ddot{U} = \partial T_{12}/\partial b$$
$$\rho\ddot{U} = \partial T_{22}/\partial b, \text{ and} \qquad (39)$$
$$\rho\ddot{W} = \partial T_{32}/\partial b \,.$$

Differentiating (38b), (38e), and (38h) we get

$$\frac{\partial T_{12}}{\partial b} = \frac{1}{2}(C_{11} - C_{12}) \cdot U_{bb}$$

$$+ (C_{11} + C_{166} + C_{266})(U_bV_{bb} + V_bU_{bb}) \qquad (40a)$$

$$\frac{\partial T_{22}}{\partial b} = C_{11}V_{bb} + (C_{11} + C_{166} + C_{266})U_bU_{bb}$$

$$+ (3C_{11} + C_{111} + 3C_{166} - C_{266})V_bV_{bb}$$

$$+ (C_{11} + C_{144} + 2C_{456})W_bW_{bb} \tag{40b}$$

$$\frac{\partial T_{32}}{\partial b} = C_{44} \cdot W_{bb} + (C_{11} + C_{144} + 2C_{456})(V_bW_{bb} + W_bV_{bb}) \tag{40c}$$

For longitudinal waves in this direction, we have U = 0 and W = 0. So the equations of motion reduce to

$$\rho_0 \frac{\partial^2 V}{\partial t^2} = C_{11} \frac{\partial^2 V}{\partial b^2}$$

$$+ (3C_{11} + C_{111} + 3C_{166} - C_{266}) \frac{\partial V}{\partial b} \quad \frac{\partial^2 V}{\partial b^2} \tag{41}$$

or

$$\rho_0 \frac{\partial^2 V}{\partial t^2} - \alpha \frac{\partial^2 V}{\partial b^2} = \delta \frac{\partial V}{\partial b} \cdot \frac{\partial^2 V}{\partial b^2} \tag{42}$$

where

$$\alpha = C_{11}$$

and

$$\delta = 3C_{11} + C_{111} + 3C_{166} - C_{266}$$

C. <u>Longitudinal Wave Propagation along the [001] Direction (or the C-Axis)</u>

For this special case the only nonvanishing displacement components are

$$U = U(c,t), \quad V = V(c,t), \quad W = W(c,t)$$

U_a, U_b, V_a, V_b, W_a and W_b vanish. To the T_{ij}'s for this case reduce to

$$T_{11} = C_{13} \cdot W_c + (\tfrac{1}{2} C_{13} + C_{44} + \tfrac{1}{2} C_{144} + C_{456})U_c^2$$
$$+ (\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{144} - C_{456})V_c^2 + (\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{133})W_c^2 \quad (43a)$$

$$T_{12} = (C_{44} + 2C_{456})U_c V_c \quad (43b)$$

$$T_{13} = C_{44} \cdot U_c + (C_{33} + C_{344})U_c W_c \quad (43c)$$

$$T_{21} = (C_{44} + 2C_{456})U_c V_c \quad (43d)$$

$$T_{22} = C_{13} \cdot W_c + (\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{144} - C_{456})U_c^2$$
$$+ (\tfrac{1}{2} C_{13} + C_{44} + \tfrac{1}{2} C_{144} + C_{456})V_c^2$$
$$+ (\tfrac{1}{2} C_{13} + \tfrac{1}{2} C_{133})W_c^2 \quad (43e)$$

$$T_{23} = C_{44}V_c + (C_{33} + C_{344})V_c W_c \quad (43f)$$

$$T_{31} = C_{44}U_c + (C_{44} + C_{344})U_c W_c \quad (43g)$$

$$T_{32} = C_{44}V_c + (C_{44} + C_{344})V_c W_c \quad (43h)$$

$$T_{33} = C_{33}W_c + (\tfrac{1}{2} C_{33} + \tfrac{1}{2} C_{344})U_c^2$$
$$+ (\tfrac{1}{2} C_{33} + \tfrac{1}{2} C_{344})V_c^2 + (\tfrac{3}{2} C_{33} + \tfrac{1}{2} C_{333})W_c^2 \quad (43i)$$

The equations of motion for plane wave propagation in the [001] direction are

$$\begin{aligned}
\rho_0 \ddot{U} &= \partial T_{13}/\partial c \\
\rho_0 \ddot{V} &= \partial T_{23}/\partial c \\
\rho_0 \ddot{W} &= \partial T_{33}/\partial c
\end{aligned} \tag{44}$$

Differentiating (43c), (43f) and (43i) with respect to c, we get

$$\partial T_{13}/\partial c = C_{44}U_{cc} + (C_{33} + C_{344})(U_cW_{cc} + W_cU_{cc}) \tag{45a}$$

$$\partial T_{23}/\partial c = C_{44}V_{cc} + (C_{33} + C_{344})(V_cW_{cc} + W_cV_{cc}) \tag{45b}$$

$$\begin{aligned}
\partial T_{33}/\partial c = C_{33}W_{cc} &+ (C_{33} + C_{344})U_cU_{cc} \\
&+ (C_{33} + C_{344})V_cV_{cc} \\
&+ (3C_{33} + C_{333})W_cW_{cc}
\end{aligned} \tag{45c}$$

For longitudinal waves in the c-direction, U = 0 and V = 0. So the only equation of motion that does not vanish is

$$\rho_0 \frac{\partial^2 W}{\partial t^2} = C_{33} \frac{\partial^2 W}{\partial c^2} + (3C_{33} + C_{333}) \frac{\partial W}{\partial c} \frac{\partial^2 W}{\partial c^2} \tag{46}$$

or

$$\frac{\partial^2 W}{\partial t^2} - C_{33} \frac{\partial^2 W}{\partial c^2} = (3C_{33} + C_{333}) \frac{\partial W}{\partial c} \frac{\partial^2 W}{\partial c^2} \tag{47}$$

which can be rewritten as

$$\rho \frac{\partial^2 W}{\partial t^2} - \alpha \frac{\partial^2 W}{\partial c^2} = \delta \frac{\partial W}{\partial c} \frac{\partial^2 W}{\partial c^2} \tag{48}$$

where

$$\alpha = C_{33}$$

and

$$\delta = 3C_{33} + C_{333} .$$

D. Solution to the Equations of Motion

The equations of motion for longitudinal waves propagating along the [100], [010], and [001] directions given by Eqs. (37), (42), and (48) have the same general form given by

$$\rho_0 \ddot{U} - \alpha U_{aa} = \delta U_a U_{aa} . \tag{49}$$

So we see that a pure mode longitudinal wave may propagate in a nonlinear hexagonal solid along the three directions [100], [010], and [001]. As other workers have noticed for cubic crystals [20], the nonlinear term causes the wave to generate harmonics as it propagates. In order to solve Eq. (49), a perturbation approach is necessary. Let us try the solution

$$U = U_1 + U_2 . \tag{50}$$

Here U_1 and U_2 represent, respectively, the approximate solutions of first and second order of the equation. By substituting Eq. (50) into Eq. (49) one gets

$$\rho_0 \ddot{U}_1 + \rho_0 \ddot{U}_2 - \alpha U_{1aa} - \alpha U_{2aa} = \delta(U_{1a} + U_{2a})(U_{1aa} + U_{2aa}) \tag{51}$$

Keeping only first order terms leads to the linear wave equation:

$$\rho_0 \ddot{U}_1 - \alpha U_{1aa} = 0 , \tag{52}$$

where $(\alpha/\rho_0)^{1/2} = C_0$ is the phase velocity of the linear wave. Substituting (52) into (51), we have

$$\rho_0 \ddot{U}_2 - \alpha U_{2aa} = \delta[U_{1a}U_{1aa} + (U_{2a}U_{1aa} + U_{1a}U_{2aa}) + U_{2a}U_{2aa}] \tag{53}$$

Since $U_2 << U_1$, the second and third terms on the right hand side are very small compared to the first term. Thus, to a second approximation the solution is reduced to

$$\rho_0 \ddot{U}_2 - \alpha U_{2aa} = \delta U_{1a}U_{1aa} . \tag{54}$$

We impose the boundary condition that

$$\left.\begin{aligned} U_1 &= A \sin \omega t \\ U_2 &= 0 \end{aligned} \qquad \text{where } a = 0 \right\} \tag{55}$$

Thus, a solution for the linear equation (52) which satisfies the boundary condition is

$$U_1 = A \sin(ka - \omega t) . \tag{56}$$

Now, substituting (56) into (54), we may rewrite as follows:

$$\rho_0 \ddot{U}_2 - \alpha U_{2aa} = - \frac{1}{2} \delta k^3A^2 \sin 2(ka - \omega t) . \tag{57}$$

Taking into account the boundary condition (55) for U_2, we use a trial solution in the following form:

$$U_2 = B\ a \sin 2(ka - \omega t) + C\ a \cos 2(ka - \omega t) \tag{58}$$

where B and C are coefficients to be determined. Putting the solution into (57), we get

$$4(-\omega^2\rho_0 Ba + k^2\alpha Ba + k\alpha C)\sin 2(ka - \omega t) +$$

$$4(-\omega^2\rho_0 Ca - k\alpha B + k^2\alpha CA)\cos 2(ka - \omega t) = -\frac{\delta k^3 A^2}{2}\sin^2(ka - \omega t)\ .$$

Now, equating the coefficients on both sides of the equation we get

$$\left.\begin{array}{l} -\omega^2\rho_0 Ba + k^2\alpha Ba + k\alpha C = -\frac{1}{8}\delta k^3 A^2 \\ \text{and} \\ -\omega^2\rho_0 C - k\alpha B + k^2\alpha Ca = 0 \end{array}\right\} \tag{59}$$

But

$$\alpha = {}_0C_0{}^2$$

and

$$\omega = C_0 k\ ,$$

so Eqs. (59) become

$$-C_0{}^2k^2\rho_0 Ba + k^2\rho_0 C_0{}^2 Ba + k\rho_0 C_0{}^2 C = -\frac{\delta}{8}k^3A^2 \tag{59a}$$

and

$$C_0{}^2k^2\rho_0 Ca - k\rho_0 C_0{}^2 B + k^2\rho_0 C_0{}^2 Ca = 0\ . \tag{59b}$$

Dividing Eq. (59a) by $k\rho_0 C_0{}^2$, we get

$$C = -\frac{\delta(kA)^2}{\delta\rho_0 C_0{}^2}\ . \tag{60a}$$

Also, dividing Eq. (59b) by $k\rho C_0^2$, we get

$$B = 0 \ . \tag{60b}$$

So, after one iteration the solution to the equation of motion becomes

$$U(a,t) = A \sin(ka - \omega t) - \left[\frac{\delta (kA)^2}{8\rho_0 C_0^2}\right] a \cos 2(ka - \omega t) \ . \tag{61}$$

The equation of wave propagation in the nonlinear regime as given in Eq. (49) was

$$\rho_0 \frac{d^2u}{dt^2} - \alpha \frac{d^2u}{da^2} = \delta \frac{du}{da} \cdot \frac{d^2u}{da^2} \tag{62}$$

Breazeale and Ford[20] write the same equation from the fluid analogy (for cubic crystals) as

$$\rho_0 \frac{d^2u}{dt^2} = K_2\left(\frac{d^2u}{da^2} + 3 \frac{du}{da} \cdot \frac{d^2u}{da^2}\right) + K_3 \frac{du}{da} \cdot \frac{d^2u}{da^2}$$

or

$$\rho_0 \frac{d^2u}{dt^2} - K_2 \frac{d^2u}{da^2} = (3K_2 + K_3) \frac{du}{da} \cdot \frac{d^2u}{da^2} \ . \tag{63}$$

Equations (62) and (63) are the same if we put

$$\alpha = K_2$$

and (64)

$$\delta = (3K_2 + K_3)$$

Now, if we consider the linear wave equation (Eq. 52) we can write it as

$$\rho_0 \frac{d^2u}{dt^2} = \alpha \frac{d^2u}{da^2}$$

or

$$\frac{d^2u}{dt^2} = \left(\frac{\alpha}{\rho_0}\right) \frac{d^2u}{da^2} . \qquad (65)$$

Comparing this with the linear wave equation

$$\frac{d^2u}{dt^2} = C_0^2 \frac{d^2u}{da^2} \qquad (66)$$

in which C_0 is the phase velocity (for small-amplitude waves), one sees that

$$K_2 = \alpha = \rho_0 C_0^2 . \qquad (67)$$

These substitutions allow us to write the solution to the nonlinear wave equation (Eq. 61) as:

$$u(a,t) = A \sin(ka - \omega t) - \left[\frac{\delta}{8\alpha}\right](kA)^2 a \cos 2(ka - \omega t) \qquad (68)$$

in complete analogy to cubic crystals; however, the expressions for α and δ, or alternatively K_2 and K_3 must be examined for hexagonal symmetry.

E. The Ultrasonic Nonlinearity Parameter for Hexagonal Crystals

As in the case of cubic crystals [1], we can define the ultrasonic nonlinearity parameter for hexagonal crystals as the negative of the ratio of the nonlinear term to the linear term in the nonlinear wave equation [Eq. (63)]. It is given by

$$\beta = -\frac{\delta}{\alpha} . \tag{69}$$

For an initially sinusoidal disturbance at a = 0, the solutions as given by Eq. (68) can be written in terms of the nonlinearity parameter as

$$u = A_1 \sin(ka - \omega t) + \frac{A_1^2 k^2 a}{8} \beta \cos 2(ka - \omega t) \tag{70}$$

where A_1 is the amplitude of the fundamental wave and $A_2 = \frac{A_1^2 k^2 a}{8} \beta$ is the amplitude of the generated second harmonic wave. In terms of A_1 and A_2, is given by

$$\beta = 8\left(\frac{A_2}{A_1^2}\right) \frac{1}{k^2 a} . \tag{71}$$

So by measuring A_1 and A_2 one can determine β which can be used to evaluate the δ's which are combinations of SOE and TOE constants, or the K_3's which are combinations of TOE constants only. The parameters α (or K_2), δ (or $(3K_2 + K_3)$) and K_3 (or $(\delta - 3K_2)$) for the three symmetry directions considered in Sections A, B, and C given by Eqs. (37), (42), and (48) are written in a tabular form in Table I. Note that C_{166} and C_{266} are combinations of TOE constants (given by

Table I. The K_2 and K_3 Parameters for Hexagonal Crystals along the Directions Considered in Sections A, B, and C

Direction of Wave Propagation	$K_2 = \alpha$	$K_3 = (\delta - 3\alpha)$
[100] or a-axis	C_{11}	$(C_{111} + 3C_{166} + C_{266})$ or $(2C_{222} - C_{112})$
[010] or b-axis	C_{11}	$(C_{111} + 3C_{166} - C_{266})$ or $(\frac{5}{2} C_{222} - C_{111} - \frac{1}{2} C_{112})$
[001] or c-axis	C_{33}	C_{333}

Eqs. (26)). In Table I we have given the K_3 parameters in terms of these combinations as well as in terms of the independent TOE constants.

F. The Equations of Motion when the Coordinate System is Rotated by 60° about the c-axis

In the remaining calculations we will consider the nonlinear equation of motion for various directions in the basal plane. As was shown by Borgnis[6] and Brugger,[7] any direction in the basal plane is a pure mode direction in the linear approximation. By determining the nonlinear equation of motion for several directions we can gain insight about the way the nonlinearity parameter changes as the propagation direction changes in the basal plane.

In order to derive the equations of motion for waves generating along the a'-axis, we have to derive the expression for strain energy in the frame of reference in which the a-axis in the original frame is rotated by 60° about the c-axis. Consider the propagation of plane finite amplitude waves along the a'-axis. Rotate the basal plane in the original frame by 60° or about the c-axis. So the coordinate frame formation can be written as

$$\begin{vmatrix} a' \\ b' \\ c' \end{vmatrix} = (R) \begin{vmatrix} a \\ b \\ c \end{vmatrix} \tag{72}$$

where the rotation matrix is given by

$$(R) = \begin{vmatrix} \cos\theta & \sin\theta & 0 \\ -\sin\theta & \cos\theta & 0 \\ 0 & 0 & 1 \end{vmatrix} \tag{73}$$

where θ is the angle through which the original basal plane has been rotated about the c-axis. We rotate the a-axis by $\theta = 60°$ in which case

$$(R) = \begin{vmatrix} 1/2 & \sqrt{3/2} & 0 \\ -\sqrt{3/2} & 1/2 & 0 \\ 0 & 0 & 1 \end{vmatrix} \tag{74}$$

Let us define the displacement components in the new coordinate system as

$$\begin{aligned} u' &= u'\,(a',t) \\ v' &= v'\,(a',t) \\ w' &= w'\,(a',t) \end{aligned} \tag{75}$$

etc.

Using the transformation matrix as given by Eq. (74) one can transform the strain components by [9]

$$(\eta) = (R^*)(\eta')(R)\ , \tag{76}$$

where

$$R^* = \begin{vmatrix} 1/2 & -\sqrt{3/2} & 0 \\ \sqrt{3/2} & 1/2 & 0 \\ 0 & 0 & 1 \end{vmatrix} \tag{77}$$

to arrive at the following expression:

$$\eta = \begin{vmatrix} 1/2 & -\sqrt{3}/2 & 0 \\ \sqrt{3}/2 & 1/2 & 0 \\ 0 & 0 & 1 \end{vmatrix} \begin{vmatrix} \eta'_{11} & \eta'_{12} & \eta'_{13} \\ \eta'_{21} & \eta'_{22} & \eta'_{23} \\ \eta'_{31} & \eta'_{32} & \eta'_{33} \end{vmatrix} \begin{vmatrix} 1/2 & \sqrt{3}/2 & 0 \\ -\sqrt{3}/2 & 1/2 & 0 \\ 0 & 0 & 1 \end{vmatrix} \tag{78}$$

Multiplication and simplification lead to the following expressions for the strain components in the original frame of reference expressed in terms of the strain components in the rotated or primed coordinate system.

$$\begin{aligned}
\eta_{11} &= \frac{1}{4}\eta'_{11} - \frac{\sqrt{3}}{4}\eta'_{12} - \frac{\sqrt{3}}{4}\eta'_{21} + \frac{3}{4}\eta'_{22} \\
\eta_{12} &= \frac{\sqrt{3}}{4}\eta'_{11} + \frac{1}{4}\eta'_{12} - \frac{3}{4}\eta'_{21} - \frac{\sqrt{3}}{4}\eta'_{22} \\
\eta_{13} &= \frac{1}{2}\eta'_{13} - \frac{\sqrt{3}}{2}\eta'_{23} \\
\eta_{21} &= \frac{\sqrt{3}}{4}\eta'_{11} - \frac{3}{4}\eta'_{12} + \frac{1}{4}\eta'_{21} - \frac{\sqrt{3}}{4}\eta'_{22} \\
\eta_{22} &= \frac{3}{4}\eta'_{11} + \frac{\sqrt{3}}{4}\eta'_{12} + \frac{\sqrt{3}}{4}\eta'_{21} + \frac{1}{4}\eta'_{22} \qquad (79) \\
\eta_{23} &= \frac{\sqrt{3}}{2}\eta'_{13} + \frac{1}{2}\eta'_{23} \\
\eta_{31} &= \frac{1}{2}\eta'_{31} - \frac{\sqrt{3}}{2}\eta'_{32} \\
\eta_{32} &= \frac{\sqrt{3}}{2}\eta'_{31} + \frac{1}{2}\eta'_{32} \\
\eta_{33} &= \eta'_{33}
\end{aligned}$$

Substitution of Eq. (79) in Eq. (25) gives the following expression for the strain energy in the primed coordinate system.

$$
\begin{aligned}
\phi' = {} & \frac{1}{2} C_{11}\{(\eta'_{11})^2 + (\eta'_{22})^2 + (\eta'_{12})^2 + (\eta'_{21})^2\} \\
& + C_{12}\{\eta'_{11}\eta'_{22} - \frac{1}{2}(\eta'_{12})^2 - \frac{1}{2}(\eta'_{21})^2\} \\
& + C_{13}\{\eta'_{11}\eta'_{33} + \eta'_{22}\eta'_{33}\} + \frac{1}{2} C_{33}(\eta'_{33})^2 \\
& + C_{44}\ \{(\eta'_{13}) + (\eta'_{31})^2 + (\eta'_{23})^2 + (\eta'_{32})^2\} \\
& + \frac{1}{6} C_{111}\{(\eta'_{11})^3 + 3(\eta'_{11})^2\eta'_{22} + 3(\eta'_{22})^2\eta'_{11} + (\eta'_{22})^3\} \\
& + \frac{1}{2} C_{113}\eta'_{33}\{(\eta'_{11})^2 + 2\eta'_{11}\eta'_{22} + (\eta'_{22})^2\} \\
& + \frac{1}{2} C_{133}\{(\eta'_{33})^2\eta'_{11} + (\eta'_{33})^2\eta'_{22}\} \\
& + C_{144}(\eta'_{11} + \eta'_{22})\{(\eta'_{23})^2 + (\eta'_{32})^2 + (\eta'_{13})^2 + (\eta'_{31})^2\} \\
& + \frac{1}{2} C_{166}\{(\eta'_{11})^3 + (\eta'_{22})^3 + 2(\eta'_{12})^2\eta'_{11} \\
& + 2(\eta'_{21})^2\eta'_{11} - (\eta'_{22})^2\eta'_{11} - (\eta'_{11})^2\eta'_{22} \\
& + 2(\eta'_{12})^2\eta'_{22} + 2(\eta'_{21})^2\eta'_{22}\} \\
& + \frac{1}{6} C_{266}\{(\eta'_{11})^3 + \frac{3\sqrt{3}}{2}(\eta'_{12})^3 + \frac{3\sqrt{3}}{2}(\eta'_{21})^3 \\
& - (\eta'_{22})^3 - \frac{3}{2}(\eta'_{12})^2\eta'_{11} - \frac{3}{2}(\eta'_{21})^2\eta'_{11}
\end{aligned}
$$

$$+ 3(\eta_{22}')^2\eta_{11}' - 3(\eta_{11}')^2\eta_{22}' - 9\eta_{11}'\eta_{12}'\eta_{21}'$$

$$- \frac{3\sqrt{3}}{2}(\eta_{21}')^2\eta_{12}' - \frac{3\sqrt{3}}{2}(\eta_{12}')^2\eta_{21}'$$

$$+ \frac{3}{2}(\eta_{21}')^2\eta_{22}' + \frac{3}{2}(\eta_{12}')^2\eta_{22}' + 9\eta_{12}'\eta_{21}'\eta_{22}'\}$$

$$+ \frac{1}{6} C_{333}(\eta_{33}')^3$$

$$+ C_{344}\eta_{33}'\{(\eta_{23}')^2 + (\eta_{32}')^2 + (\eta_{13}')^2 + (\eta_{31}')^2\}$$

$$+ \frac{1}{2} C_{366}\eta_{33}'\{(\eta_{11}')^2 + 2(\eta_{12}')^2 + 2(\eta_{21}')^2 + (\eta_{22}')^2 - 2\eta_{11}'\eta_{22}'\}$$

$$+ \frac{1}{4} C_{456}\{[\frac{1}{2}(\eta_{11}') + \frac{\sqrt{3}}{2}\eta_{12}' + \frac{\sqrt{3}}{2}\eta_{21}' - \frac{1}{2}\eta_{22}']$$

$$\times [\frac{1}{2}(\eta_{13}')^2 - \frac{1}{2}(\eta_{23}')^2 + \sqrt{3}\,\eta_{13}'\eta_{23}' + \frac{1}{2}(\eta_{31}')^2$$

$$- \frac{1}{2}(\eta_{32}')^2 + \sqrt{3}(\eta_{31}'\eta_{32}')]$$

$$+ \eta_{13}'\eta_{31}'[\frac{3}{2}(\eta_{11}') - \frac{\sqrt{3}}{2}(\eta_{12}') - \frac{\sqrt{3}}{2}\eta_{21}' - \frac{3}{2}\eta_{22}']$$

$$+ \eta_{23}'\eta_{31}'[-\frac{\sqrt{3}}{2}(\eta_{11}') + \frac{5}{2}(\eta_{12}') - \frac{3}{2}\eta_{21}' + \frac{\sqrt{3}}{2}\eta_{22}']$$

$$+ \eta_{32}'\eta_{13}'[-\frac{\sqrt{3}}{2}\eta_{11}' - \frac{3}{2}\eta_{12}' + \frac{5}{2}\eta_{21}' + \frac{\sqrt{3}}{2}\eta_{22}']$$

$$+ \eta_{32}'\eta_{23}'[-\frac{3}{2}\eta_{11}' + \frac{\sqrt{3}}{2}\eta_{12}' + \frac{\sqrt{3}}{2}\eta_{21}' + \frac{3}{2}\eta_{22}']\} \tag{80}$$

Once the expression for strain energy is obtained, one can execute the algebra exactly as in Sections A, B, and C and derive the equations

of motion for plane pure mode longitudinal waves propagating along the [100], [010], and [001] symmetry axes in the rotated frame of reference. This has been done and we found that for these directions also the equation of motion has the same form as Eq. (49) and its solution follows exactly as in Section D. The parameters α and δ appearing in the equation of motion and the corresponding K_2 and K_3 parameters are tabulated in Table II.

From Tables I and II one can notice that the a-axis and a'-axis or the [100] direction in the original and rotated coordinate systems are equivalent. Similarly, the directions perpendicular to them which are the [010] directions in the two cases are also equivalent. Of course, the c-axis is the same as before; e.g., the rotation operation is performed about the c-axis.

G. The Equations of Motion when the Coordinate System is Rotated by $\pi/4$ about the c-Axis

In order to derive the equations of motion for the propagation of plane longitudinal waves along the [110] direction in the basal plane, we have to derive the expression for strain energy in the frame of reference in which the a-axis in the original coordinate system is rotated by 45° about the c-axis. The coordinate transformation for this case is:

$$\begin{vmatrix} a' \\ b' \\ c' \end{vmatrix} = (R) \begin{vmatrix} a \\ b \\ c \end{vmatrix} \tag{81}$$

Table II. The K_2 and K_3 Parameters for Hexagonal Crystals in the Rotated Frame of Reference Described in Section F

Direction of Wave Propagation	$K_2 = \alpha$	$K_3 = (\delta - 3\alpha)$
[100] or the a'-axis	C_{11}	$(C_{111} + 3C_{166} + C_{266})$ or $(2C_{222} - C_{112})$
[010] or the b'-axis	C_{11}	$(C_{111} + 3C_{166} - C_{266})$ or $(\frac{5}{2} C_{222} - C_{111} - \frac{1}{2} C_{112})$
[001] or the c-axis	C_{33}	C_{333}

where

$$R = \begin{vmatrix} 1/\sqrt{2} & 1/\sqrt{2} & 0 \\ -1/\sqrt{2} & 1/\sqrt{2} & 0 \\ 0 & 0 & 1 \end{vmatrix} \tag{82}$$

is the transformation matrix from Eq. (73). The components of strain in the initial frame can be expressed in terms of the components of strain in the rotated frame as

$$\eta = R^* \, (\eta') \, R \tag{83}$$

$$\begin{vmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} \end{vmatrix} = \begin{vmatrix} 1/\sqrt{2} & -1/\sqrt{2} & 0 \\ 1/\sqrt{2} & 1/\sqrt{2} & 0 \\ 0 & 0 & 1 \end{vmatrix} \begin{vmatrix} \eta'_{11} & \eta'_{12} & \eta'_{13} \\ \eta'_{21} & \eta'_{22} & \eta'_{23} \\ \eta'_{31} & \eta'_{32} & \eta'_{33} \end{vmatrix} \begin{vmatrix} 1/\sqrt{2} & 1/\sqrt{2} & 0 \\ -1/\sqrt{2} & 1/\sqrt{2} & 0 \\ 0 & 0 & 0 \end{vmatrix} \tag{84}$$

Expansion and simplification gives the following expressions for η_{ij}'s in terms of η_{ij}'s.

$$\eta_{11} = \frac{1}{2}\left(\eta'_{11} - \eta'_{12} - \eta'_{21} + \eta'_{22}\right)$$

$$\eta_{12} = \frac{1}{2}\left(\eta'_{11} + \eta'_{12} - \eta'_{21} - \eta'_{22}\right)$$

$$\eta_{13} = \frac{1}{\sqrt{2}}\left(\eta'_{13} - \eta'_{23}\right)$$

$$\eta_{21} = \frac{1}{2}\left(\eta'_{11} - \eta'_{12} + \eta'_{21} - \eta'_{22}\right)$$

$$\eta_{22} = \frac{1}{2}\left(\eta'_{11} + \eta'_{12} + \eta'_{21} + \eta'_{22}\right)$$

$$\eta_{23} = \frac{1}{\sqrt{2}} (\eta'_{13} + \eta'_{23})$$

$$\eta_{31} = \frac{1}{\sqrt{2}} (\eta'_{31} - \eta'_{32})$$

$$\eta_{32} = \frac{1}{\sqrt{2}} (\eta'_{31} + \eta'_{32})$$

$$\eta_{33} = \eta'_{33} \tag{85}$$

Substituting Eq. (85) into Eq. (25), we get the following expression for the elastic strain energy in the rotated or primed frame of reference.

$$\begin{aligned}
\phi' = \frac{1}{2} C_{11}[\eta'^{2}_{11} + \eta'^{2}_{22} + \eta'^{2}_{12} + \eta'^{2}_{21}] \\
+ C_{12}(\eta'_{11}\eta'_{22} - \frac{1}{2} \eta'^{2}_{12} - \frac{1}{2} \eta'^{2}_{21}] \\
+ C_{13}[\eta'_{11}\eta'_{33} + \eta'_{22}\eta'_{33}] + \frac{1}{2} C_{33}\eta'^{2}_{33} \\
+ C_{44}[\eta'^{2}_{13} + \eta'^{2}_{31} + \eta'^{2}_{23} + \eta'^{2}_{32}] + \frac{1}{6} C_{333}\eta'^{3}_{33} \\
+ \frac{1}{6} C_{111}[\eta'^{3}_{11} + 3\eta'^{2}_{11} \eta'_{22} + 3\eta'^{2}_{22}\eta'_{11} + \eta'^{3}_{22}] \\
+ \frac{1}{2} C_{133}\eta'^{2}_{33}[\eta'_{11} + \eta'_{22}] \\
+ \frac{1}{2} C_{113} \eta'_{33}[\eta'^{2}_{11} + 2\eta'_{11}\eta'_{22} + \eta'^{2}_{22}] \\
+ C_{144}(\eta'_{11} + \eta'_{22})[\eta'^{2}_{23} + \eta'^{2}_{32} + \eta'^{2}_{13} + \eta'^{2}_{31}] \\
+ C_{344}\eta'_{33}[\eta'^{2}_{23} + \eta'^{2}_{32} + \eta'^{2}_{13} + \eta'^{2}_{31}]
\end{aligned}$$

$$+ \frac{1}{2} C_{166}(\eta'_{11} + \eta'_{22})[\eta'^{2}_{11} + \eta'^{2}_{22} + 2\eta'^{2}_{12} + 2\eta'^{2}_{21} - 2\eta'_{11}\eta'_{22}]$$

$$+ \frac{1}{6} C_{266}[2\eta'^{3}_{12} + 2\eta'^{3}_{21} - 6\eta'^{2}_{12}\eta_{21} - 6\eta'^{2}_{21}\eta_{12}$$

$$+ 3\eta'^{2}_{11}\eta'_{12} + 3\eta'^{2}_{11}\eta'_{21} + 3\eta'^{2}_{22}\eta'_{12} + 3\eta'^{2}_{22}\eta'_{21}$$

$$- 6\eta'_{11}\eta'_{12}\eta'_{22} - 6\eta'_{11}\eta'_{21}\eta'_{22}]$$

$$+ \frac{1}{2} C_{366}\eta'_{33}[\eta'^{2}_{11} + 2\eta'^{2}_{12} + 2\eta'^{2}_{21} + \eta'^{2}_{22} - 2\eta'_{11}\eta'_{22}]$$

$$+ 2C_{456}[2(\eta'_{12} + \eta'_{21})(\eta'_{13}\eta'_{23} + \eta'_{31}\eta'_{32})$$

$$+ 2(\eta'_{13}\eta'_{31} - \eta'_{23}\eta'_{32})(\eta'_{11} - \eta'_{22})$$

$$+ 2(\eta'_{13}\eta'_{32} - \eta'_{23}\eta'_{31})(\eta'_{21} - \eta'_{12})] \tag{86}$$

The derivatives of the strain energy with respect to the strain components are obtained by taking the approximate derivatives of Eq. (86). Then, substituting them in Eqs. (20), one obtains the components of stress tensor. These stress components are differentiated with respect to the appropriate displacement to obtain the equations of motion for longitudinal waves propagating along [100], [010], and [001] directions in the rotated frame which are, respectively, the [110], [$\bar{1}$10], and [001] directions in the original coordinate system. The algebra is similar to that performed in Sections A, B, and C, and is not reproduced here.

It is found that for these directions also, the equations of motion have the same form as Eq. (49) and the solution follows as in

Section D. So it is found that a pure mode longitudinal waves propagate along these directions with the generation of harmonics. The parameters α and δ appearing in the equations of motion and the corresponding K_2 and K_3 parameters are tabulated in Table III.

An examination of Table III reveals that for the [100] and [010] directions in the 45° rotated frame, the K_2 and K_3 values are the same. This can be expected from the symmetry of hexagonal crystals. Obviously, the values for the c-axis do not change, because the rotation was about the c-axis. The K_2 values are the same, i.e., C_{11}, for every direction in the basal plane. This is consistent with the fact that the velocity of longitudinal waves is the same ($V = \sqrt{C_{11}/\rho_0}$) for every direction in the basal plane.

H. <u>Summary of Results</u>

We can arrive at some very important conclusions by comparing Tables I, II, and III. They predict the relative magnitudes of the second harmonic generated when waves are propagated along different directions in a hexagonal crystal. The K_3 parameters for a-axis and a'-axis are the same, i.e., $(2C_{222} - C_{112})$. The K_3 values for directions perpendicular to them (the [010] direction) are also the same, i.e., $(5/2\ C_{222} - C_{111} - 1/2\ C_{112})$. These directions make a 30° angle with the a- and b-axes. By symmetry the 30°, 90°, and 150° directions are equivalent and they have the same K_3 parameter. The K_3 values for the [110] direction and [$\bar{1}$10] directions which make a 45° angle with the a-axis (and 15° with the b-axis), as given in Table III,

Table III. The K_2 and K_3 Parameters for Hexagonal Crystals in the Rotated Frame of Reference Described in Section G

Direction of Wave Propagation	$K_2 = \alpha$	$K_3 = (\delta - 3\alpha)$
[100] or the [110] in the Original Frame	C_{11}	$C_{111} + 3C_{166}$ or $\frac{1}{4}(9C_{222} - 3C_{112} - 2C_{111})$
[010] or the [$\bar{1}$10] in the Original Frame	C_{11}	$C_{111} + 3C_{166}$ or $\frac{1}{4}(9C_{222} - 3C_{112} - 2C_{111})$
[001] or the c-Axis	C_{33}	C_{333}

are the same, i.e., $\frac{1}{4}(9C_{222} - 3C_{112} - 2C_{111})$. By symmetry the directions which make 15°, 45°, 75°, 105°, etc. are equivalent and they have the same K_3 parameter. Taking into account the symmetry of different directions we have combined Tables I, II, and III to make Table IV, which is a summary of the results.

Another important observation that can be made is that the difference in K_3 parameters between that for the a-axis and that for the 45° direction is the same as the difference between the K_3 parameters for the 45° direction (or 15° direction) and the 30° direction. The value of the difference is C_{266} or $\frac{1}{4}(2C_{111} - C_{112} - C_{222})$. Note that always the angles of the various directions are measured from the a-axis which is taken as the [100] direction. The average of the K_3 parameters for [100] and 30° direction is the K_3 parameter for 15° direction (or 45° direction). This suggests a diagram similar to the slowness diagrams of Musgrave,[5] as shown in Figure 2. However, in Figure 2 we indicate the magnitude of the K_3 parameter in the basal plane. The slowness diagram for longitudinal waves in the basal plane of a hexagonal crystal would be a circle because the velocity is the same in all directions. Even though the velocity is the same in all directions, the amplitude of the generated second harmonic varies with direction, as indicated in Figure 2. This means that by measuring the second harmonic and calculating K_3 along the [100], the [110] and the [010] directions in a hexagonal crystal one should be able to isolate C_{266} and the combination $(C_{111} + 3C_{166})$, or equivalently, the combinations $(2C_{222} - C_{112})$ and $\frac{1}{2}(3C_{112} - 4C_{111})$. Measuring K_3 along the c-axis should give C_{333}.

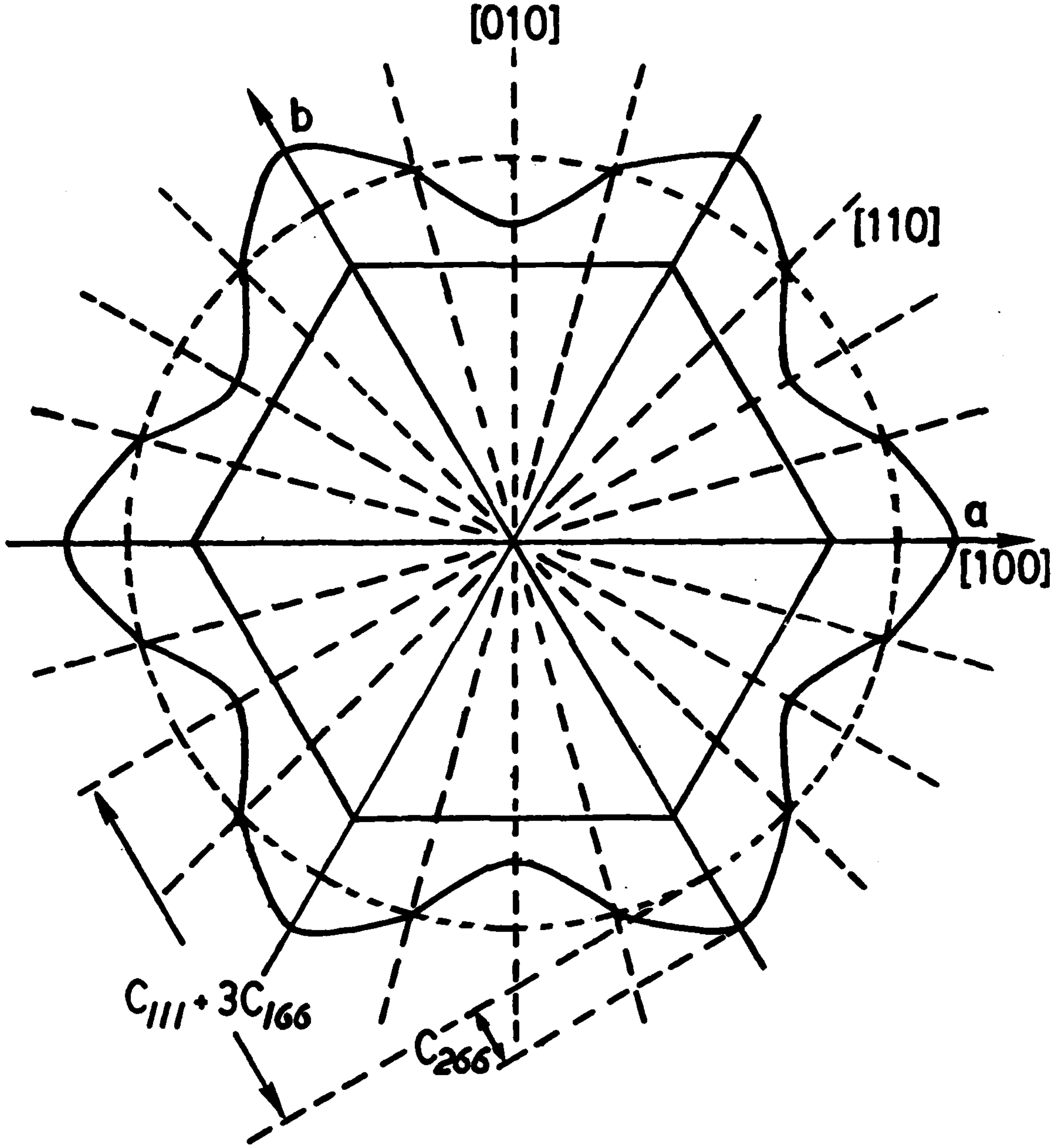


Fig. 2. Magnitude of the K_3 parameter plotted as a function of angle in the basal plane.

Table IV. The K_2 and K_3 Parameters for Hexagonal Crystals (Summary of Tables I, II, and III)

Direction of Wave Propagation	K_2	K_3
[100], the a-Axis or a'-Axis	C_{11}	$(C_{111} + 3C_{166} + C_{266})$ or $(2C_{222} - C_{112})$
[010], 30° or 90° Direction to a-Axis	C_{11}	$(C_{111} + 3C_{166} - C_{266})$ or $\frac{1}{2}[5C_{222} - 2C_{111} - C_{112}]$
[110], 15°, 45° or 75° Direction to a-Axis	C_{11}	$(C_{111} + 3C_{166})$ or $\frac{1}{4}[9C_{222} - 3C_{112} - 2C_{111}]$
[001], the c-Axis	C_{33}	C_{333}

Finally, according to our interpretation of the figure giving the pure mode directions in a hexagonal crystal in the publication of Brugger,[15] there remains a set of pure mode directions for which we have not yet been able to derive a nonlinear theory. These are the directions along a cone whose apex angle is centered about the c-axis. Such a derivation may be contained in a later technical report; however, at present it appears that the theory may become so complicated that one can derive the results only for specific crystals rather than in general terms as given above for the directions we have considered.

REFERENCES

1. Jacob Philip, Technical Report No. 19, Ultrasonics Group, Department of Physics, University of Tennessee, Knoxville (AD A097724).

2. R. F. S. Hearmon, in Landolt-Bornstein Numerical Data and Functional Relationships in Science and Technology, edited by K. H. Hellwege (Springer-Verlag, Berlin, Heidelberg, New York, 1979), Vol. 11.

3. M. J. P. Musgrave, Proc. Roy. Sco. A 226, 356 (1954).

4. G. W. Farnell, Can. J. Phys. 39, 65 (1961).

5. M. J. P. Musgrave, Crystal Acoustics (Holden Day, San Francisco, 1970).

6. F. E. Borgnis, Phys. Rev. 98, 1000 (1955).

7. K. Brugger, J. Appl. Phys. 36, 759 (1965).

8. Z. P. Chang, J. Appl. Phys. 39, 5669 (1968).

9. F. D. Murnaghan, Finite Deformations of an Elastic Solid (Wiley, New York, 1951).

10. F. Birch, Phys. Rev. 71, 809 (1947).

11. A. Seeger and O. Buck, Z. Naturforsch. 15a, 1056 (1960).

12. N. G. Einspruch and R. J. Manning, J. Appl. Phys. 35, 560 (1964).

13. R. N. Thurston and K. Brugger, Phys. Rev. 133, A1604 (1964).

14. R. N. Thurston, in Physical Acoustics, edited by W. P. Mason (Academic Press, New York, 1964), Vol. I A.

15. K. Brugger, J. Appl. Phys. 36, 768 (1965).

16. K. Brugger, Phys. Rev. 133, A1611 (1964).

17. D. Lazarus, Phys. Rev. 76, 545 (1949).

18. D. S. Hughes and J. L. Kelly, Phys. Rev. 92, 1145 (1953).

19. T. Bateman, W. P. Mason, and H. J. McSkimin, J. Appl. Phys. 32, 928 (1961).

20. M. A. Breazeale and J. Ford, J. Appl. Phys. 36, 3486 (1965).

21. M. A. Breazeale and J. Philip, in Physical Acoustics, edited by W. P. Mason (Academic Press, New York, in press).

22. R. E. Green, Jr., in Treatise on Materials Science and Technology (Academic Press, New York, 1973), Vol. 3.

23. K. D. Swartz and C. Elbaum, Phys. Rev. B 1, 1512 (1970).

24. E. R. Naimon, Phys. Rev. B 4, 4291 (1971).

25. D. J. Silversmith and B. L. Averbach, Phys. Rev. B 1, 567 (1970).

26. E. S. Fisher, M. H. Manghnani and T. J. Sokolowski, J. Appl. Phys. 41, 2991 (1970).

27. E. S. Fisher, M. H. Manghnani and R. Kikuta, J. Phys. Chem. Solids 34, 687 (1973).

28. M. H. Manghnani, K. Katahara and E. S. Fisher, Phys. Rev. B 9, 1421 (1974).

29. R. E. Schmunk and C. S. Smith, J. Phys. Chem. Solids 9, 100 (1959).

30. J. A. Corll, ONR Technical Report No. 6 (1962) [unpublished].

31. E. S. Fisher and M. H. Manghnani, J. Phys. Chem. Sclids 32, 657 (1971).

32. R. Ramji Rao and A. Ramanand, Phys. Stat. Solidi 58a, 11 (1980).

33. Z. A. Goldberg, Sov. Phys.-Acoustics 6, 306 (1961).

REPORTS DISTRIBUTION LIST FOR ONR PHYSICS DIVISION OFFICE
UNCLASSIFIED CONTRACTS

Director Defense Advanced Research Projects Agency Attn: Technical Library 1400 Wilson Blvd. Arlington, Virginia 22209	1 copy
Office of Naval Research Physics Division Office (Code 412) 800 North Quincy Street Arlington, Virginia 22217	2 copies
Office of Naval Research Director, Technology (Code 200) 800 North Quincy Street Arlington, Virginia 22217	1 copy
Naval Research Laboratory Department of the Navy Attn: Technical Library Washington, DC 20375	1 copy
Office of the Director of Defense Research and Engineering Information Office Library Branch The Pentagon Washington, DC 20301	1 copy
U.S. Army Research Office Box 1211 Research Triangle Park North Carolina 27709	2 copies
Defense Technical Information Center Cameron Station Alexandria, Virginia 22314	12 copies
Director, National Bureau of Standards Attn: Technical Library Washington, DC 20234	1 copy
Director U.S. Army Engineering Research and Development Laboratories Attn: Technical Documents Center Fort Belvoir, Virginia 22060	1 copy
ODDR&E Advisory Group on Electron Devices 201 Varick Street New York, New York 10014	1 copy

Air Force Office of Scientific Research Department of the Air Force Bolling AFB, DC 22209	1 copy
Air Force Weapons Laboratory Technical Library Kirtland Air Force Base Albuquerque, New Mexico 87117	1 copy
Air Force Avionics Laboratory Air Force Systems Command Technical Library Wright-Patterson Air Force Base Dayton, Ohio 45433	1 copy
Lawrence Livermore Laboratory Attn: Dr. W. F. Krupke University of California P.O. Box 808 Livermore, California 94550	1 copy
Harry Diamond Laboratories Technical Library 2800 Powder Mill Road Adelphi, Maryland 20783	1 copy
Naval Air Development Center Attn: Technical Library Johnsville Warminster, Pennsylvania 18974	1 copy
Naval Weapons Center Technical Library (Code 753) China Lake, California 93555	1 copy
Naval Underwater Systems Center Technical Center New London, Connecticut 06320	1 copy
Commandant of the Marine Corps Scientific Advisor (Code RD-1) Washington, DC 20380	1 copy
Naval Ordnance Station Technical Library Indian Head, Maryland 20640	1 copy
Naval Postgraduate School Technical Library (Code 0212) Monterey, California 93940	1 copy
Naval Missile Center Technical Library (Code 5632.2) Point Mugu, California 93010	1 copy

Naval Ordnance Station Technical Library Louisville, Kentucky 40214	1 copy
Commanding Officer Naval Ocean Research & Development Activity Technical Library NSTL Station, Mississippi 39529	1 copy
Naval Explosive Ordnance Disposal Facility Technical Library Indian Head, Maryland 20640	1 copy
Naval Ocean Systems Center Technical Library San Diego, California 92152	1 copy
Naval Surface Weapons Center Technical Library Silver Spring, Maryland 20910	1 copy
Naval Ship Research and Development Center Central Library (Code L42 and L43) Bethesda, Maryland 20084	1 copy
Naval Avionics Facility Technical Library Indianapolis, Indiana 46218	1 copy
Dr. Bill D. Cook Dept. of Mechanical Engineering University of Houston Houston, Texas 77004	1 copy
Dr. Floyd Dunn Biophysical Research Laboratory University of Illinois Urbana, Illinois 61801	1 copy
Dr. E. F. Carome Department of Physics John Carroll University University Heights Cleveland, Ohio 44017	1 copy
Albert Goldstein, Ph.D. Dept. of Radiology Harper-Grace Hospitals 3990 John R. Detroit, MI 48201	1 copy

END

FILMED

11-84

DTIC